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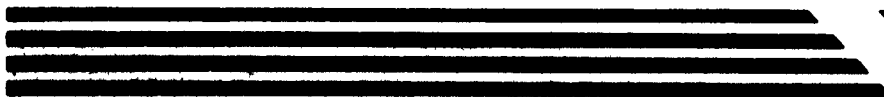
ARMY MEDICAL RESEARCH LABORATORY

FORT KNOX, KENTUCKY

Report No. 89
11 August 1952

SCINTILLATION COUNTERS

*Subtask under Effects of Irradiation, AMRL Project No. ^b8-59-08-013



MEDICAL RESEARCH AND DEVELOPMENT BOARD
OFFICE OF THE SURGEON GENERAL
DEPARTMENT OF THE ARMY

REPORT NO. 89

SCINTILLATION COUNTERS*

by

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FORT KNOX, KENTUCKY
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MEDEA

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ABSTRACT

SCINTILLATION COUNTERS

OBJECT

This report is essentially a critical review of work done on scintillation counters, emphasizing radiobiological applications.

RESULTS

Since the first use of a scintillation counter in 1941, this technique has reached an extremely high state of perfection. It has opened new possibilities in research, brought important knowledge to nuclear physics, cosmic ray research and radiobiology and has led to newer theories of the mechanism of energy transport in irradiated solutions. Application of these theories to biological systems may give information for the further understanding of the protective mechanism afforded by different substances, such as cysteine and glutathione, in total body x-irradiation. The newer ideas of a simple "shielding effect" by the administered substances can be interpreted more clearly with the facts gained from scintillation research.

CONCLUSIONS

Scintillation counters have become important tools in radiobiological research and clinical investigations. Greater progress is possible, especially in the direction of an understanding of the fundamental processes involved in irradiated biological systems.

RECOMMENDATIONS

Systematic studies with scintillation counters on biological systems should be started. Special attention should be given to questions of theoretical radiobiology.

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SCINTILLATION COUNTERS

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SCINTILLATION COUNTERS

I. HISTORICAL SKETCH

In 1903 Elster and Geitel (1) studied the luminescence of zinc-sulfide under alpha-ray bombardment with a magnifier and a microscope. The microscope showed the luminescence to consist of single individual lightflashes, the "scintillations". Crookes (2), who studied this phenomenon in the same year constructed the "spinthariscopes" for the subjective observation of the scintillations.

In 1908 Regener (3) proved that each individual alpha particle produced an individual scintillation. He developed a method based on this principle for the counting of alpha-rays. This method was highly welcomed in radioactive research and was used extensively, especially by the English group (4) and the Vienna group (5). For more than two decades it was one of the most powerful tools in nuclear research and led to many important discoveries (6). The last of the great discoveries achieved with this method was the first man-made transmutation of atoms, the successful disintegration of lithium with artificially accelerated protons by Cockcroft and Walton (7) in 1932.

In spite of its advantages, however, the method was neither ideal nor convenient. Disadvantages were numerous. The scintillations, being very weak, could be observed only in the dark with well rested and well adapted eyes. The danger of undesirable gamma-ray exposure during the observations required special measures and experimental setups. Fatigue and subjective influences made a frequent change of observers necessary. They could only count for widely spaced intervals of one and a half minutes totaling not more than two hours counting per week. This counting procedure was very inefficient since reliable countings were possible only if there were as few as 20 to 40 scintillations per minute.

Thus, the method was discarded following the discovery and development of the more reliable Geiger point counter and the Geiger-Mueller counter in the nineteen twenties. The Geiger-Mueller counter, in particular, became for many years the most sensitive instrument of modern research.

A new development became apparent with the possibility of objectively measuring scintillations by using a rapidly responding highly sensitive photoelectric device in place of the eye. In 1941 Krebs (8) demonstrated for the first time the realization of this idea. Using

the Geiger photon counter, one of the most sensitive photoelectrical devices, he recorded with an oscilloscope, loudspeaker and electrical counter the scintillations produced by polonium alpha-rays in ZnS. To demonstrate the practical application of this scintillation arrangement he determined the diffusion velocity of radium emanation and thorium emanation in glass tubes.

After World War II this principle of detecting and studying scintillations with a highly sensitive photoelectric device was applied and advanced by Blau and Dreyfus (9), Coltman and Marshall (10), Sherr (11), Kallmann (12), Broser and Kallmann (13), Mandeville and Albrecht (14), Mandeville and Scherb (15), and others.

It "heralded", as Pringle (16) writes, a new area of scientific development and research, being according to Morton and Mitchell (17) one of the most important advances in devices for the detection of nuclear radiations since the invention of the Geiger-Mueller counter in 1926. Today, modern science possesses in the scintillation counter an extremely powerful instrument which in a surprisingly short time has led to many important discoveries in nuclear physics, cosmic ray research, and radiobiology.

II. PRINCIPLE

The principle of counting scintillations with the eye is demonstrated in Figure 1, using one of Rutherford's classical arrangements. R is the radioactive preparation which emits alpha-rays. These alpha-rays hit the atoms of the gas in the chamber K. If this gas is nitrogen, H-particles are produced in the nitrogen disintegration process $N^{14}(\alpha, p)O^{17}$. These H-particles hit the scintillation screen Z producing scintillations. The scintillations are observed and counted with the microscope M, the magnification being 20 to 50 x.

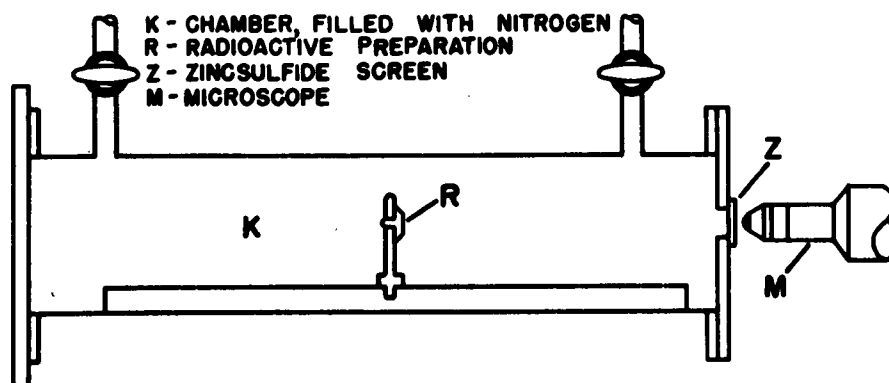


FIGURE 1.-EXPERIMENTAL ARRANGEMENT FOR THE DISINTEGRATION OF NITROGEN BY RUTHERFORD, 1919.

Under these conditions the intensity of the scintillations is, however, very weak. An alpha particle, with an energy of 5 Mev produces in a proper scintillation crystal about 10^5 quanta of the wavelength 5560 \AA . Of these 10^5 quanta only a small amount hits the eye. The duration of the scintillations is very short, for ZnS about 10^{-5} to 10^{-6} seconds. The eye can record energies of 4×10^{-7} ergs with a duration of 2×10^{-7} seconds while energies of 8×10^{-8} ergs with 8×10^{-8} seconds duration are not recorded (18). One thousand photons of the wavelength 5560 \AA have an energy of 0.36×10^{-8} ergs which is radiated in about 5×10^{-6} seconds. Hevesy and Paneth (19) maintain that the rested well adapted eye can notice a scintillation with 30 photons of the 5560 \AA range and Barnes and Czerny (20) give 40 to 90 photons as threshold values for light flashes. Scintillation experiments, however, show that at least 1200 quanta are necessary for reliable counting of scintillations (4). Alpha particles with a minimum energy of 0.13 to 0.17 Mev are not recorded according to Rutherford (4) and beta- and gamma-rays cannot be counted with this subjective method.

To demonstrate objectively, and to count automatically these scintillations, highly sensitive, rapidly responding photoelectric devices are required. Modern physics has two instruments which fulfill these requirements -- the photon counter and the photomultiplier. Both instruments have advantages and disadvantages; however, they have been used with great success for the counting and recording of scintillations.

The principle of the arrangement is the same in both cases. High energy particles or quanta hit a scintillation phosphor in which their kinetic energy is transformed into light energy and radiated in the form of light flashes, the scintillations. The scintillation light is collected by optical means on the photocathode of the detector instrument and from there channeled into the oscilloscope, amplifier, loudspeaker and recording system.

A. Photon Tube Scintillation Counter

The normal Geiger-Mueller counter has a photosensitivity of about 10^{-4} electrons per photon. This sensitivity can be increased by using proper materials for the photocathode, special arrangements of the photocathode and special treatment of sensitization (21). Electrical discharges in a hydrogen atmosphere (22) or activation in noble gas-vapor mixtures at the temperature of liquid air (23) give photon counters of high sensitivity, long lifetime and high resolution.

The scintillation device used by Krebs in 1941 is shown in Figure 2. In this case the scintillation phosphor and the photon counter are separated.

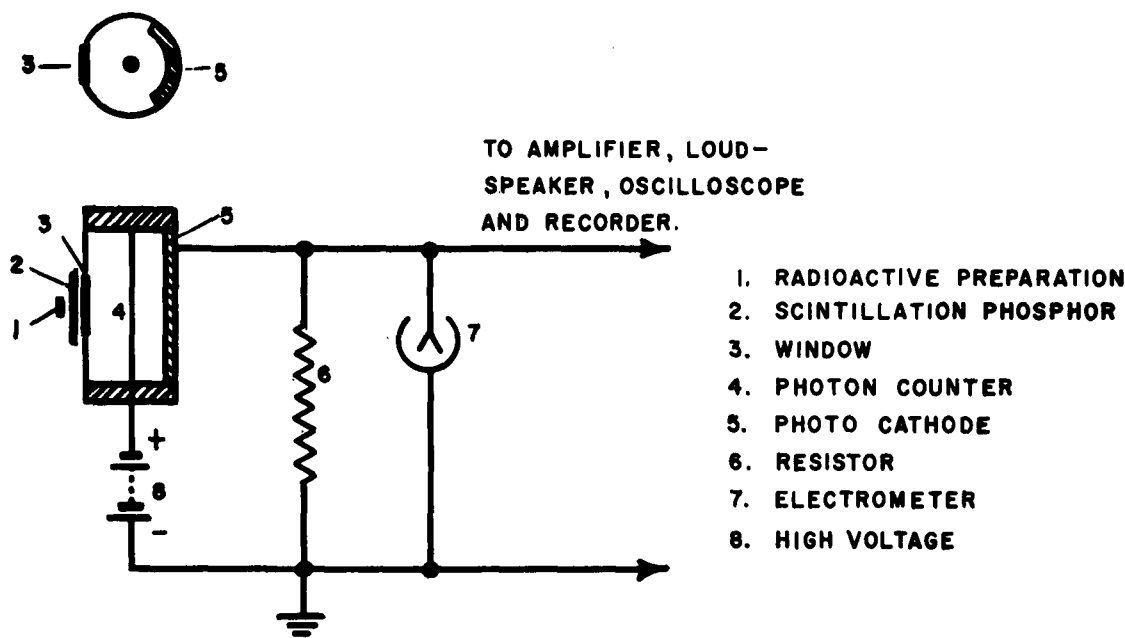


FIGURE 2.-SCINTILLATION COUNTER. KREBS, 1941.

By bringing the scintillation phosphor and the counter wall in optical contact Mandeville and Scherb (15) increased the efficiency of the scintillation counter so that not only alpha particles but also beta and gamma-rays could be counted with the device, (Figure 3).

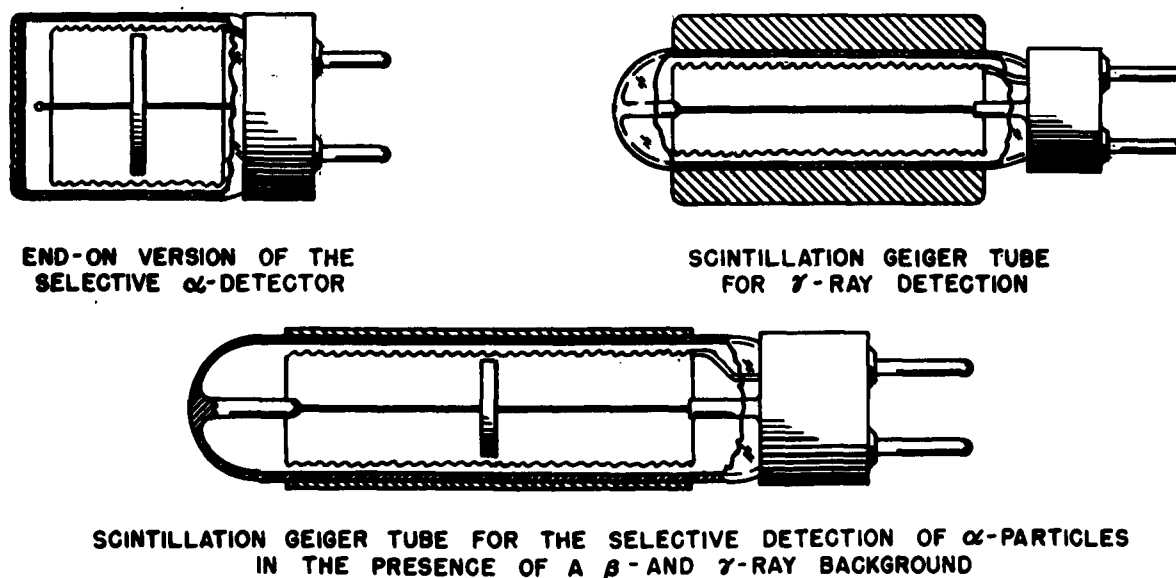
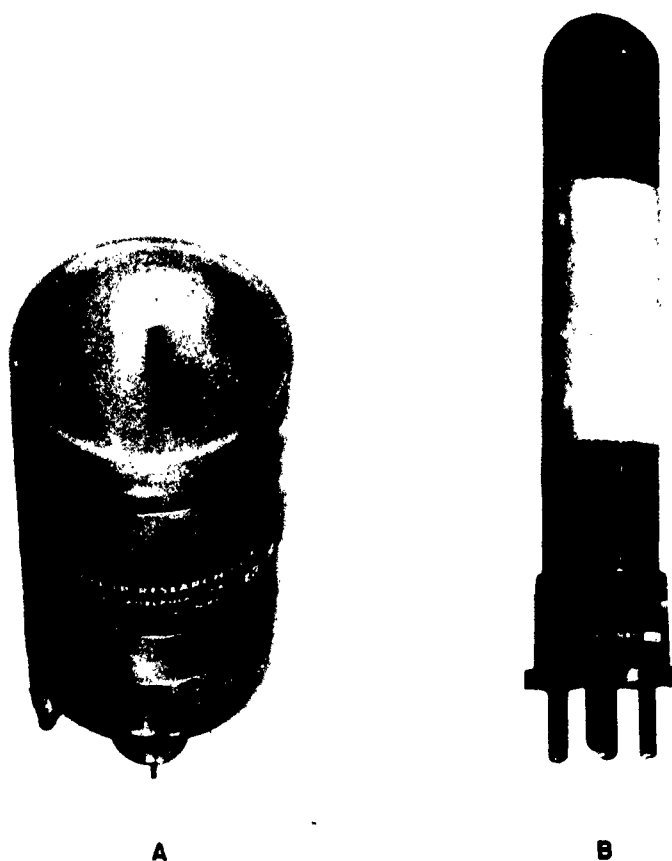


FIGURE 3.- SCINTILLATION GEIGER TUBES. MANDEVILLE AND SCHERB, 1950

To take advantage of the high resolution of the scintillation counter special circuits such as those that have been developed for the Geiger-Mueller counter had to be applied (24). The first photon tube scintillation counters worked only in the Geiger region. Devices have been developed recently (25) which count in the proportional region. Thus, this setup can be used not only for the detection of the incoming particles and quanta but also for measuring the type of radiation and its intensity. Photon tube scintillation counters offer several advantages in comparison to normal Geiger-Mueller counters. They are much more sensitive to gamma radiation and since no thin window technique is involved they also can be used for the measurement of soft radiation. Figure 4 shows two types of commercially manufactured photon tube scintillation counters.



A - TUBE NRG-S 25 A
B - TUBE NRG-S 10 A
NUCLEAR RESEARCH CORPORATION
PHILADELPHIA, PA.

FIGURE 4. SCINTILLATION GEIGER TUBES AFTER
MANDEVILLE AND SCHERB.

These counters exhibit a very high area sensitivity as shown by Daggs, Parr and Krebs (26) so that they may be applied with advantage to special problems involving small volumes.

B. Photomultiplier Scintillation Counter

The photomultiplier has been improved considerably during the last few years. The normal tubes have an amplification factor of 10^6 and more. Special tubes with a gain up to 10^{12} have been developed by Schaetti (27) and Morton (28). This high gain is reached by special construction of the tube and an increase in the number of dynodes, up to 16. Photomultipliers can be used for measuring the integral output of the scintillation light as well as for counting the single scintillations.

Figure 5 shows the arrangement of Blau and Dreyfus (9) for the measurement of the strength of radioactive preparations by the produced luminescence in a ZnS screen.

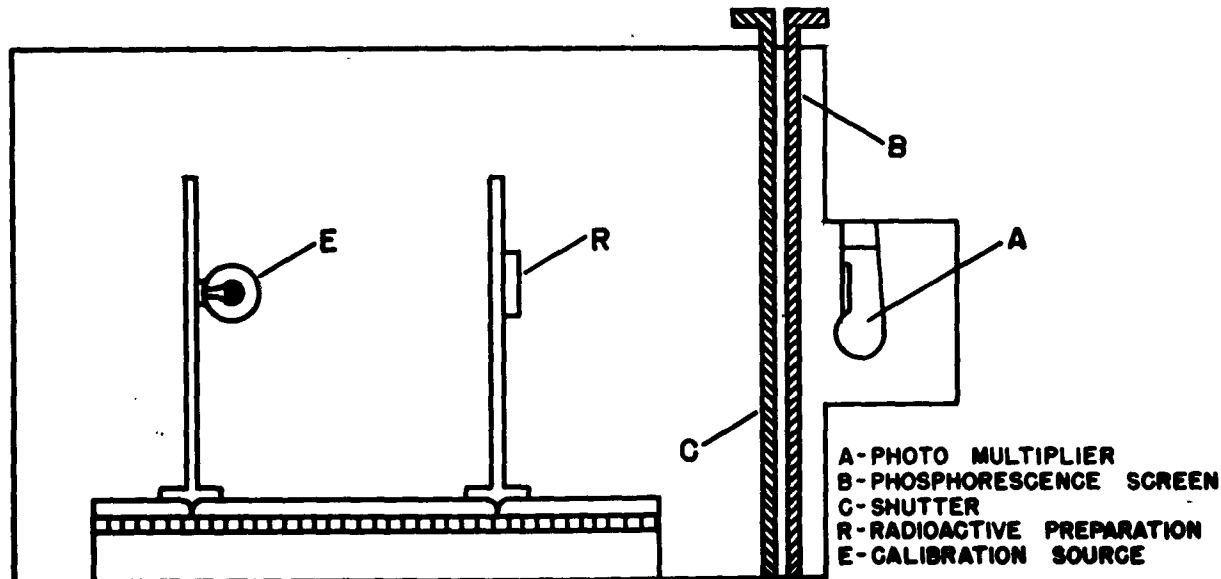


FIGURE 5.- SCINTILLATION ARRANGEMENT. BLAU AND DREYFUS, 1948

Figure 6 is a schematic illustration of the scintillation counter developed by Coltman and Marshall (10). Coltman and Marshall used a Patterson D fluorescence screen and recorded the scintillations with a RCA 931-A photomultiplier. With this arrangement they were able to measure alpha-, beta-, and gamma-rays as well as fast ions, neutrons, and x-rays.

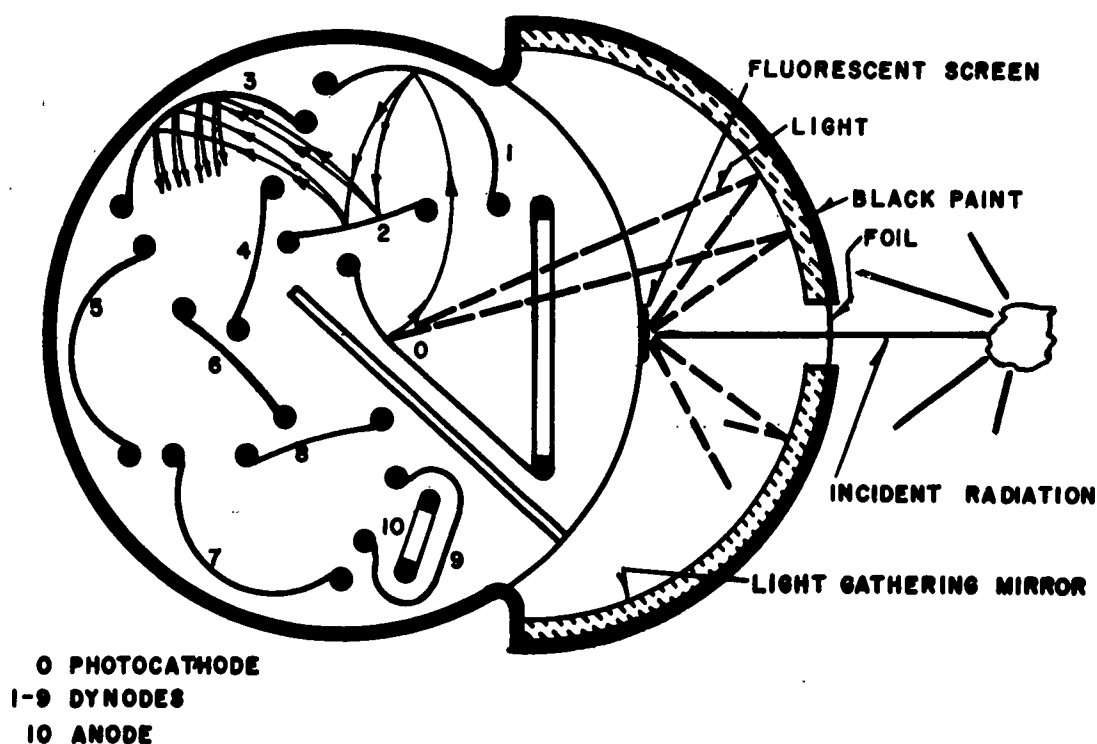


FIGURE 6.-SCHEMATIC DIAGRAM OF OPERATION OF THE SCINTILLATION COUNTER. COLTMAN AND MARSHALL, 1947

Sherr (11) described a portable alpha particle scintillation counter using a 931-A photomultiplier tube and ZnS:Ag crystals. He detected single alpha particles with a set of headphones and a greater number of alpha particles with a microammeter.

Kallmann (12), using naphthalene crystals and a photomultiplier tube, measured for the first time with great efficiency beta- and gamma-rays.

Several types of photomultipliers have proved of special value for scintillation work. They are: (a) photomultiplier RCA 931-A, a tube with a caesium-antimony photocathode the spectral response of which is given in figure 7; (b) photomultiplier 1P21, identical in construction with RCA 931-A, however with a better overall sensitivity; (c) photomultiplier 1P28, enclosed in UV transparent glass so that the spectral response reaches far into the UV; (d) photomultiplier 1P22 with a caesium-bismuth photocathode sensitive in the red region but with a lower overall sensitivity, (Figure 7).

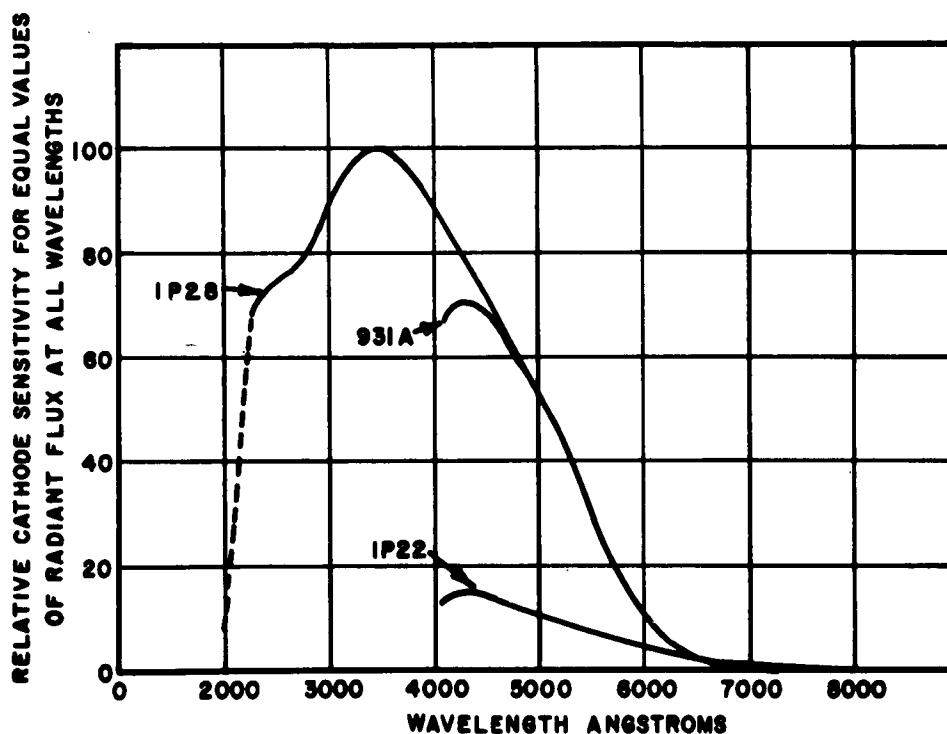


FIGURE 7.-RELATIVE CATHODE RESPONSE OF VARIOUS PHOTOMULTIPLIER TUBES AS A FUNCTION OF THE WAVELENGTH OF THE RECEIVED LIGHT.

Special data for photomultipliers 5819 and 1P21 are given in Table 1.

TABLE 1

Photomultiplier	5819	1P21
Photocathode area	11.6 cm ²	1.9 cm ²
Max. quanta efficiency	6.5 %	13.0 %
Emission max.	4800 A°	3500 A°

Good English tubes are the photomultipliers EMI 5311, EMI 5659 and VX 5031.

All photomultipliers are distinguished not only by a high gain and a high resolution but also by a high number of undesired noise pulses. These dark current noise pulses are caused by the thermal emission of electrons from the photocathode. In an oscilloscope these electrons cause a series of pulses, the amplitude of which varies according to the statistical character of the electronic amplification. For example in a selected 931-A tube there are among the 20,000 pulses with an amplitude of about 1 mm. about 15 pulses with an amplitude of 60 mm. or more. Therefore, special measures are necessary to discriminate the signal pulses from the noise pulses. Morton and Mitchell (29) showed that cooling of the photomultiplier with dry ice or liquid air greatly improved the conditions. Optical means such as reflecting mirrors, optical contact between phosphor and multiplier wall, and the wrapping of the scintillation crystal with metal foil, also support the signal pulses (30). Coincidence circuits (31) like the one developed by Kallmann and Accardo (32) permit discrimination between noise pulses and pulses of small light flashes, (Figure 8).

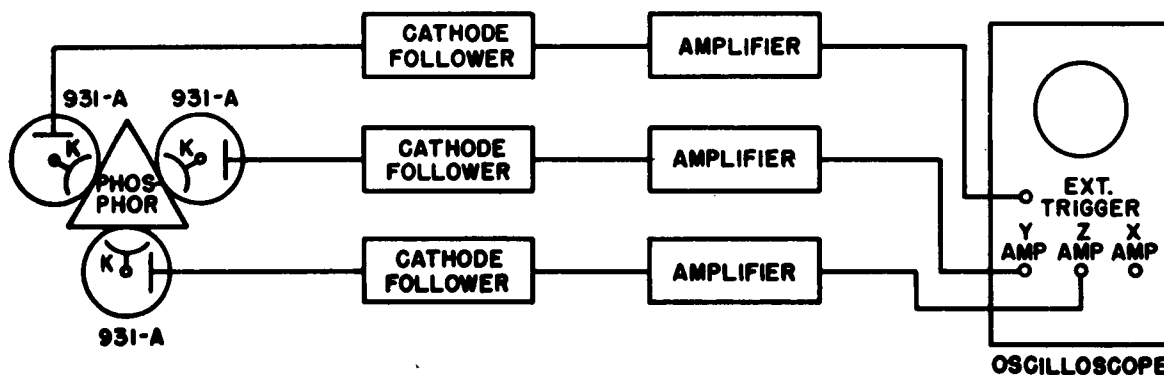
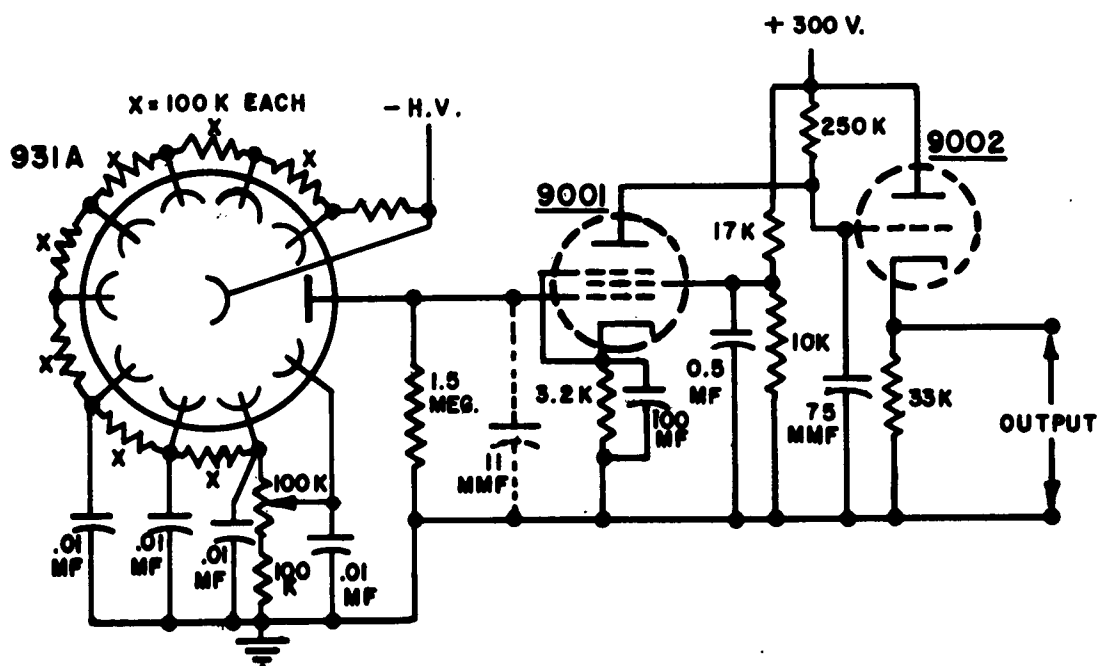


FIGURE 8.- COINCIDENCE DEVICE FOR NOISE REDUCTION, KALLMANN AND ACCARDO (32)

Figure 10 gives the diagram of the preamplifier circuit used by Coltman (33).



10

The signals of the preamplifier are fed to an amplifier, discriminator, normal scaler and an electro-mechanical counter (10, 33). By varying the bias voltage on the discriminator and taking readings at each setting the pulse height distribution of both dark current (noise pulses) and signal pulses can be obtained.

Besides the noise pulses, photomultipliers have other drawbacks, i.e. magnetic sensitivity, fatigue effects, satellite pulses, and after-pulses. The magnetic sensitivity of the tubes is so high that the response of the photomultiplier changes with different orientations in the earth's magnetic field (34). Fatigue effects of scintillation devices were reported by Blau and Dreyfus (35) as early as 1945 and have been studied systematically by Reifel, Stone and Brauner (36). To illustrate the effect, Figure 11 gives the fatigue measurement of an RCA 5819 photomultiplier tube with an anode current of 7×10^{-7} amperes. The effect does not occur as long as the total anode current is not more than 10^{-7} amperes.

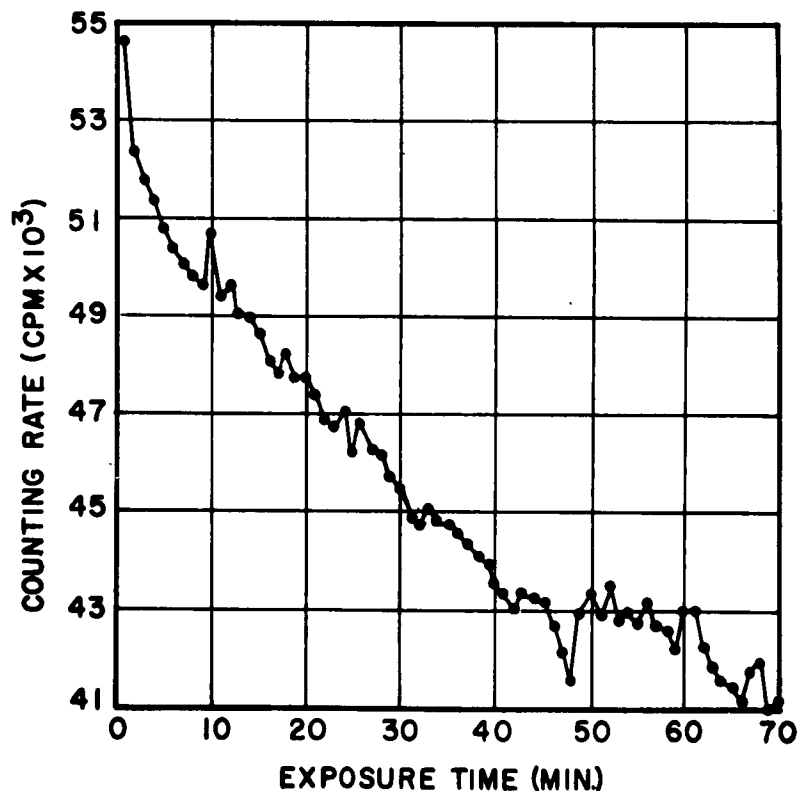


FIGURE 11.-COUNTING RATE AS A FUNCTION OF TIME FOR A 5819 TUBE WITH A 3.6-RD SOURCE AT 26 IN. AND A 1.04-MC SOURCE AT 14 IN. DISCRIMINATOR SET AT 40% AND ANODE CURRENT 7×10^{-7} AMPERES.
REIFEL, STONE AND BRAUNER (36)

Satellite pulses and afterpulsing in photomultipliers are discussed by Harrison, Godfrey and Keuffel (37) and by Mueller and co-workers (38). Satellite pulses follow the main pulse in parts of a microsecond and up to several microseconds, Mueller observing a slow component of afterpulsing up to 10 microseconds. Their amplitudes correspond to, or are slightly higher than, single electron pulses from the cathode. Satellite pulses are an inherent property of photomultipliers and they are probably caused by single positive ions formed in the photomultiplier, which in turn produce secondary electrons. The number of satellite pulses depends upon the photomultiplier voltage as demonstrated in Figure 12.

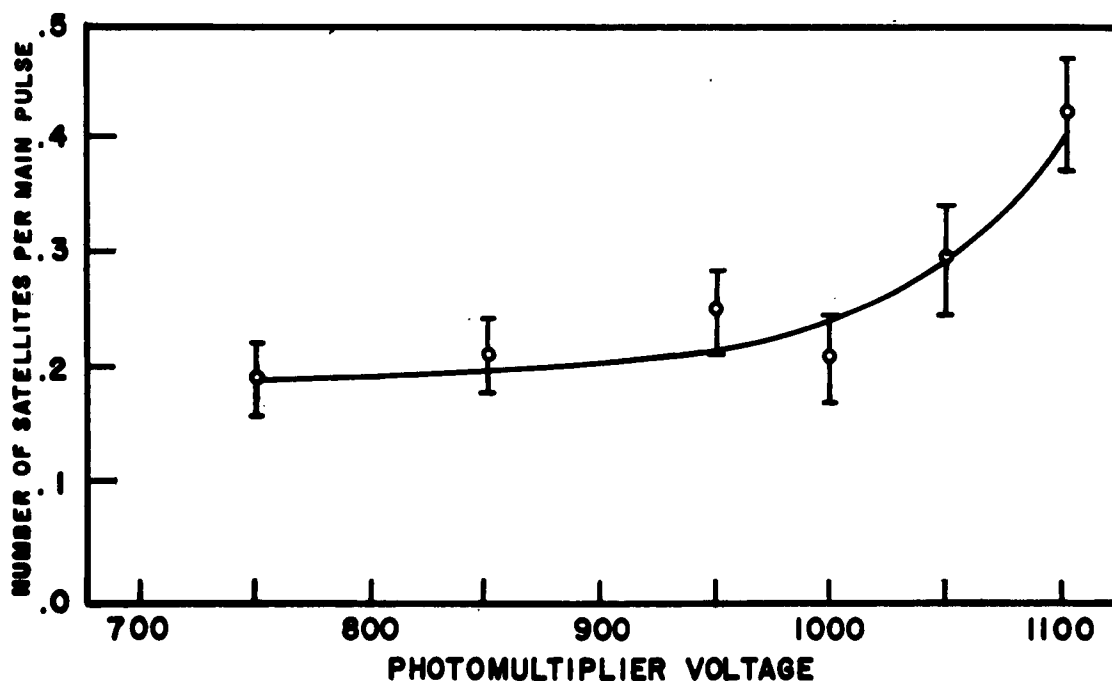


FIGURE 12.-VARIATION OF SATELLITE RATE WITH PHOTOMULTIPLIER VOLTAGE. THE ORDINATE GIVES THE NUMBER OF SATELLITES PER TRACE GREATER THAN 1/15 OF THE MINIMUM PULSE SIZE. HARRISON, GODFREY AND KEUFFEL (37)

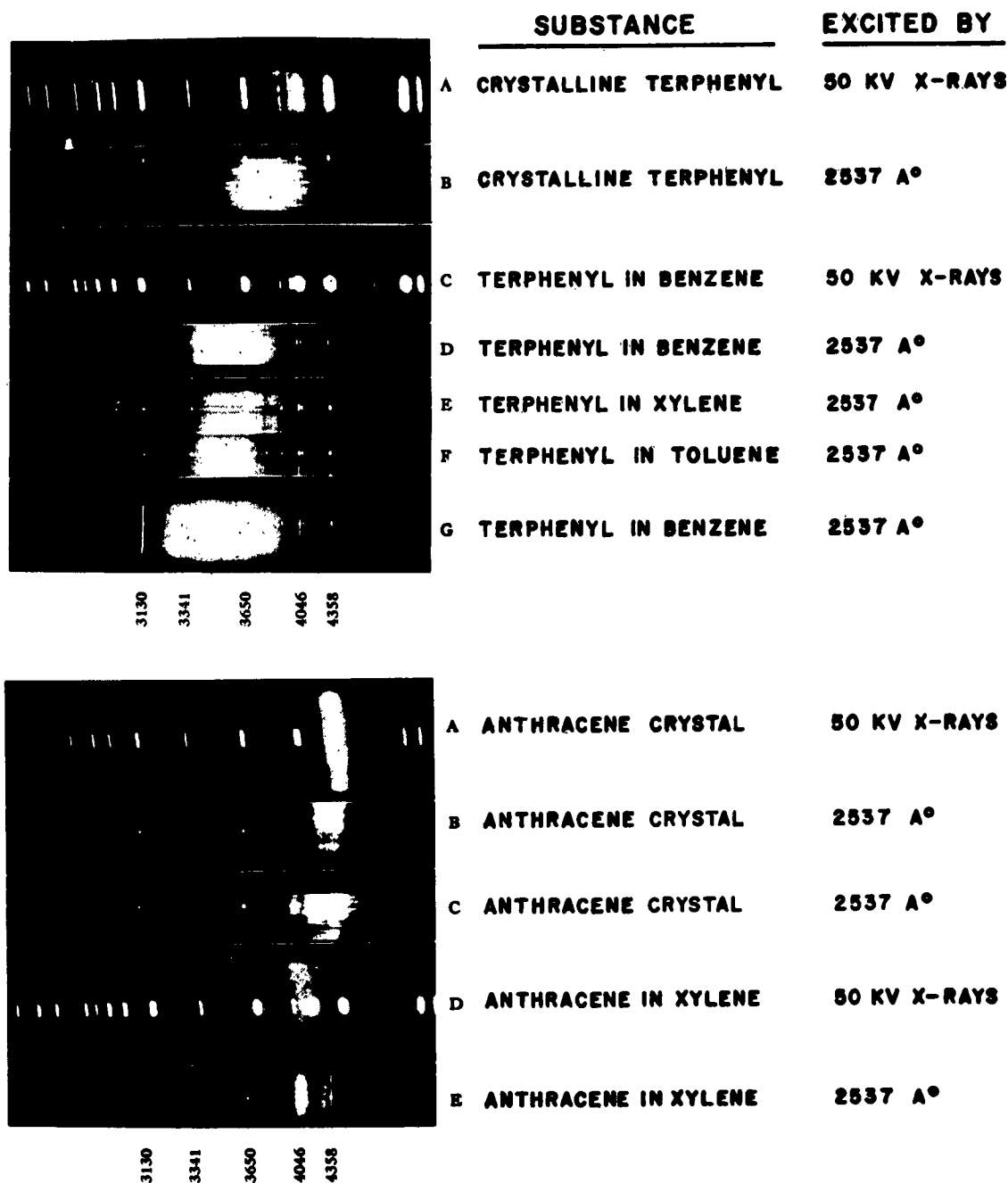


FIGURE 13. EMISSION SPECTRA OF CHARACTERISTIC ORGANIC SCINTILLATORS. HARRISON AND REYNOLDS (44).

Afterpulsing has been studied by Davison (39). It may take one or two forms. The first type starts within 10^{-8} seconds after the main pulse. The second, more complex form, shows an increase during the first microsecond and then a decrease over a period of 2 to 3 microseconds. Satellite pulses and afterpulsing present serious problems in scintillation counter experiments when short time observations and coincidence studies are to be made.

Of all factors discussed, the most important are, according to Frisch (40), the choice and the qualities of the scintillation phosphor. High scintillation efficiency, short decay time and proper optical qualities are decisive for the usefulness and the work performance of a scintillation phosphor.

III. SCINTILLATION PHOSPHORS

Zinc sulfide, Willemite, Kunzite and barium-platinum-cyanur have been the most widely used fluorescence materials. Modern scintillation research has discovered many substances, inorganic and organic, solid and liquid, pure, mixed, or in solution, which fluoresce under the influence of high energetic particles and quanta.

Improved zinc sulfides with different foreign atoms and crystals of ZnO , CaF_2 , CaWO_4 , LiAlSiO_8 , Al_2O_3 , and LiF have become important inorganic scintillation phosphors for alpha-ray counting. Valuable scintillators for beta- and gamma-ray counting have been discovered by Hofstadter (41). They are the thallium activated alkali halides, especially NaI(Tl) with 1% thallium and KI(Tl) with 0.1% thallium.

The physical efficiency of the inorganic phosphors is relatively high but their decay times are relatively long. Chariton and Lea (42) and Wood (43) showed an efficiency coefficient for alpha-rays of 0.25 and a decay time of $1/40,000$ second for the phosphor ZnS . The modern values are 0.27 for the efficiency coefficient and about 10×10^{-6} seconds for the decay time.

Excellent organic scintillators are the substances with conjugated double bonds, such as naphthalene, anthracene, terphenyl, stilbene and their mixtures. The scintillation intensities of the organic scintillators are weaker than those of ZnS but the optical transparency is higher. Therefore, large pieces can be used for beta- and gamma-ray counting. The decay times of organic phosphors are very short, the magnitude being about 10^{-8} to 10^{-9} seconds. The efficiency related to the same amount of absorbed energy, in general, is higher for beta-rays than for alpha-rays, (Table 2).

TABLE 2 *
FLUORESCENCE PROPERTIES OF SCINTILLATION SUBSTANCES
GROUP 1 - INORGANIC PHOSPHORS

SUBSTANCE	EMISSION MAX. IN Å°	DECAY CONSTANT IN 10 ⁻⁸ SECONDS	RELATIVE EFFICIENCY ANTHRACENE ± 1	ENERGY- YIELD			EFFICIENCY PER MEV	TRANSPARENCY MG/CM ²	DENSITY	MELTING POINT °C°
				α	γ	SOFT X-RAY				
ZNS:AG	4500	500	2.0	.28	.14	.20	$\eta_p = 1$	80	4.10	—
ANS:CU	5200	500	—	.25	.22	.30		200	—	—
ZNO	5500	—	—	.02	.10	.04		—	—	—
CDWO ₄	5200	600	2.0	—	—	—		—	7.90	—
CAWO ₄	4300	600	1.0	.017	.08	.04		—	6.06	—
MGWO ₄	4900	200	—	.017	—	—		100	—	—
GDS:AG	7600	1000	—	.23	—	—		HIGH	—	—
CaI ₂ (TI)	—	110	2.5	—	—	—		—	—	—
NaI(TI)	4100	25	2.0	—	—	—	$\eta_p = 1$ $\eta_{sc} = 1$	—	3.67	—
KI(TI)	4100	100	0.5	—	—	—	$\eta_p = 1$ $\eta_{sc} = 1$	—	3.13	—
KBr(TI)	3600	—	—	.017	—	—		HIGH	—	—
CSi(TI)	—	100	1.5	—	—	—		—	4.50	—

GROUP 2 ORGANIC PHOSPHORS

ANTHRA- GENE	4120	2.4	1.0	—	—	—	$\eta_p = 2$ $\eta_{sc} = 8$	—	1.25	217
	4720									
NAPHTHA- LENE	3450	6.0	0.3	.003	.05	.008		1000	1.15	80
	3850									
STILBENE	3950	0.6	0.6	—	—	—		—	1.16	124
	4200									
PHENAN- THRENE	4100	0.8	0.8	.006	.11	.015		—	1.03	100
	4300									
	4500									
TER- PHENYL	3900	0.5	0.6	—	—	—	$\eta_{sc} = 8$	—	1.23	213
	4300									
DIPHENYL	3850	1.0	0.4	.010	.075	.025		—	—	—

* REFERENCES (158-172)

TABLE 2 (CONTINUED)
GROUP 3-LIQUIDS

SUBSTANCE	RELATIVE EFFICIENCY ANTHRACENE = 1	DECAY CONSTANT IN 10^{-8} SECONDS
ANILINE	0.007	—
BENZENE	0.010	1.0
ETHYL ALCOHOL	0.009	0.1
ETHER	0.007	0.1
PARAFFIN OIL	0.014	—
STYRENE	0.006	—
TOLUENE	0.011	0.2
H ₂ O DISTILLED	0.014	—
H ₂ O DOUBLE DISTILLED	0.009	—
XYLENE	0.015	—

TABLE 2 (CONTINUED)
GROUP 4-MIXTURES AND SOLUTIONS

SUBSTANCE	SOLVENT	CONCENTRATION IN GMS/LITER	RELATIVE EFFICIENCY (γ -BOMBARD)
ANTHRACENE	BENZENE	1.0	0.066
	HEXANE	1.0	0.016
	PARAFFIN OIL	1.5	0.029
	TOLUENE	1.5	0.074
	XYLENE	1.4	0.090
NAPHTALINE	BENZENE	3.0	0.021
	XYLENE	12.0	0.025
PHENANTRENE	BENZENE	10.0	0.052
	TOLUENE	8.0	0.066
	XYLENE	7.5	0.070
TERPHENYL (p-DIPHENYL- BENZENE)	BENZENE	4.5	0.28
	HEXANE	0.8	0.049
	PARAFFIN OIL	0.5	0.045
	TOLUENE	5.0	0.39
	XYLENE	5.0	0.46
ANTHRACENE CRYSTAL	—	—	1.00

The luminescence efficiency of pure liquids is very small. The efficiency of mixtures and solutions depends upon the solvent and the solute and their mixing ratio. Mixtures and solutions in general show the properties of the added substance. In these cases, according to newer concepts, the energy of the incoming particle is absorbed by the molecules of the bulk material and from there transported by a special mechanism to the added molecules. Good solvents are phenylcyclohexane, phenylether, xylene, toluene and benzene; good solutes are anthracene, terphenyl, alpha-naphthylamine and phenyl-alpha-naphthylamine.

The decay times of liquids, solutions and mixtures also are very short, in the range of about 10^{-8} to 10^{-9} seconds. (Table 2 and Table 3).

Besides luminescence efficiency and decay time the emission spectrum of the scintillations is of importance. The emission maximum of the scintillation light should match the spectral sensitivity of the photocathode. Figure 13 shows the spectral emission of terphenyl and anthracene under UV-irradiation and X-irradiation according to Harrison and Reynolds (44). The spectrogram shows that with the use of an RCA 5819 photomultiplier, sensitivity maximum 4800 Å, the anthracene crystal meets the requirements much better than the terphenyl crystal.

TABLE 3
FLUORESCENT DECAY TIMES OF SCINTILLATION-
CRYSTALS IRRADIATED WITH UV AND GAMMA-RAYS

FLUOR	UV DECAY IN 10^{-8} SECONDS	GAMMA DECAY IN 10^{-8} SECONDS
ANTHRACENE	1.7	3.0
CARBAZOLE	0.7	1.3
DIPHENYL ACETYLENE	0.25	0.4
1,4-DIPHENYL BUTADIENE	0.2	0.6
STILBENE	0.31	0.8
TERPHENYL	1.1	0.6
0.5% TERPHENYL IN TOLUENE	0.25	0.5

The data given in the tables are corrected to zero absorption and hold for vertical incidence since the thickness of the scintillation phosphor influences the light intensity. Figure 14 shows the light emission in relation to the thickness of ZnS when bombarded with alpha-, beta-, and gamma-rays. With increasing thickness the absorption of the incoming radiation becomes more and more effective, especially in the case of alpha-rays, until the absorption of the luminescence light in the substance determines the technical efficiency of the phosphor. A ZnS thickness of 80 mg/cm^2 is the upper limit for transparency.

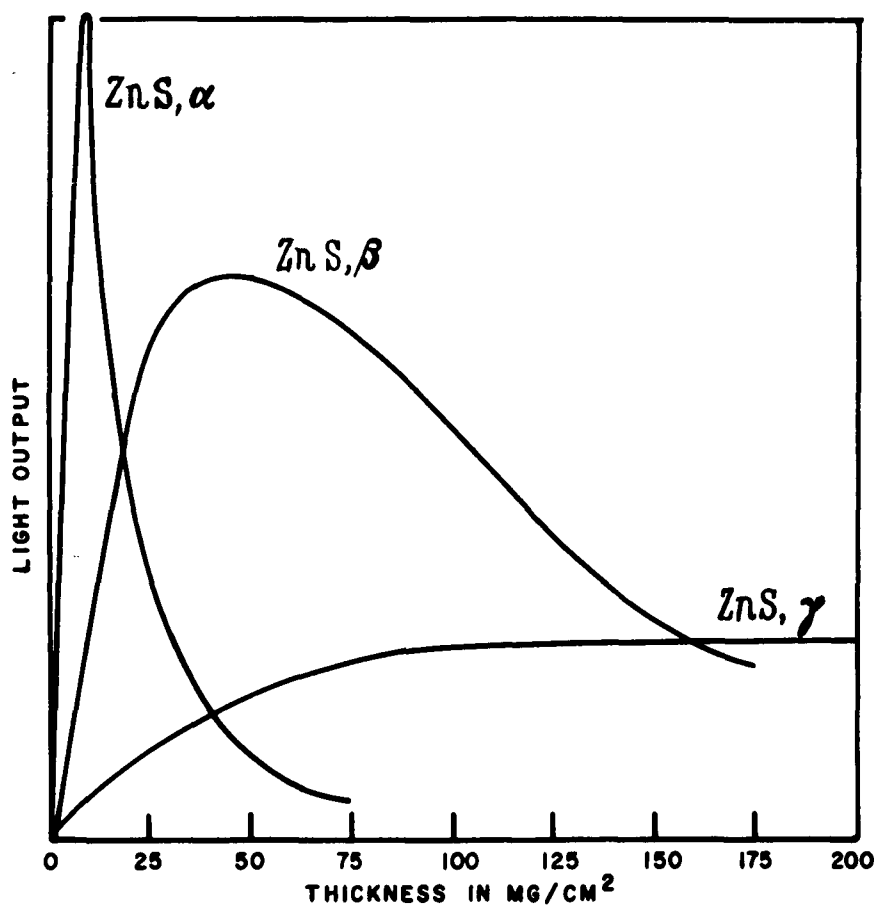


FIGURE 14.-LIGHT OUTPUT OF ZnS AS A FUNCTION OF THE THICKNESS. REPORT HANLE (172)

The exact correlation between particle energy and light output is a fundamental figure in energy measurements with scintillation counters. While in general, organic substances, both solid and liquid, do not show a linear correlation between energy and luminescence intensity, inorganic phosphors are believed to show linearity between energy and light output over a broad range. Little is known of the low energy and high energy regions. Figure 15 shows the results of measurements by Frey, Grim, Preston and Gray (45) with protons over a range from 0.4 to 17 Mev.

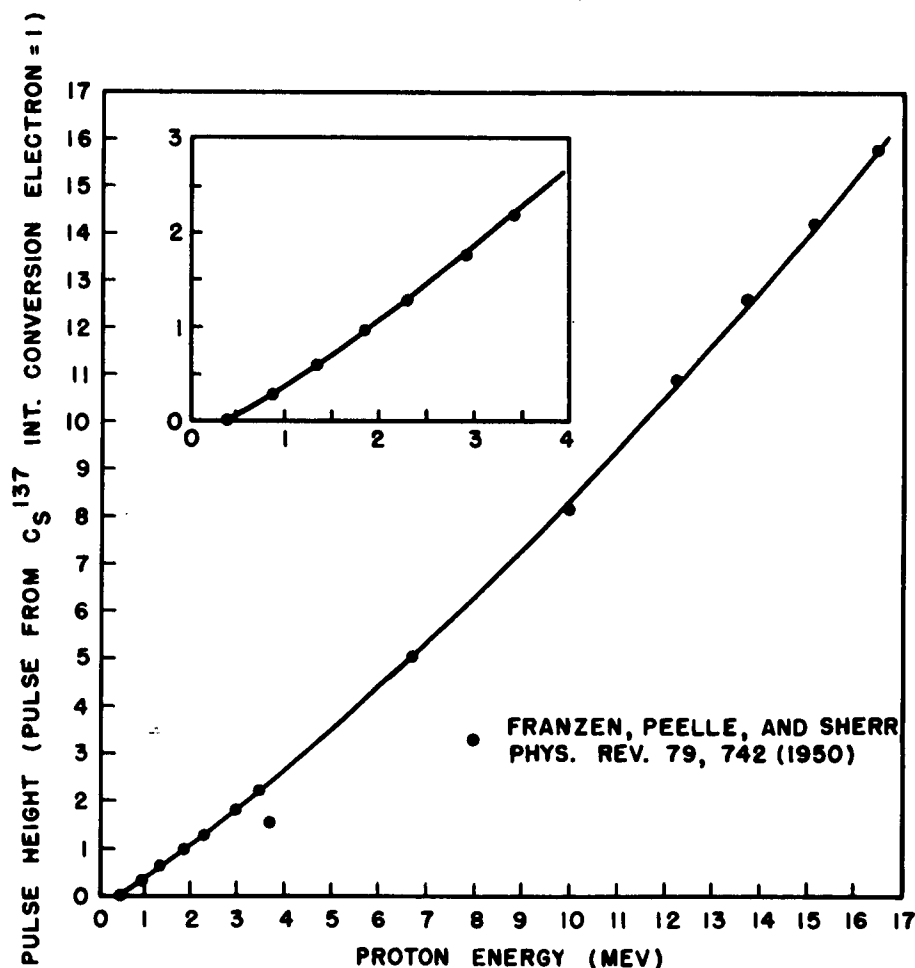


FIGURE 15.- LIGHT OUTPUT AS A FUNCTION OF PROTON ENERGY FOR AN ANTHRACENE SCINTILLATION COUNTER. THE INSERT IS AN ENLARGED PORTION SHOWING THE NEW DATA BELOW 3.7 MEV. ALTHOUGH 0.4-MEV PROTONS WERE EASILY DETECTED, THE MOST PROBABLE PULSE HEIGHT COULD NOT BE SEPARATED FROM DARK CURRENT WITH THE DATA TAKEN. FREY, GRIM, PRESTON AND GRAY (45)

Taylor, Jentschke, Remley, Eby and Kruger (46) using anthracene, stilbene and NaI(Tl) crystals studied the energy relationship for deuterons and H-ions with energies between 1 and 11 Mev; for protons with energies between 1 and 5 Mev; for alpha-rays with energies of 4 to 21 Mev; and for electrons with energies between 500 Mev and 624 kev. The experimental arrangement is shown in Figure 16.

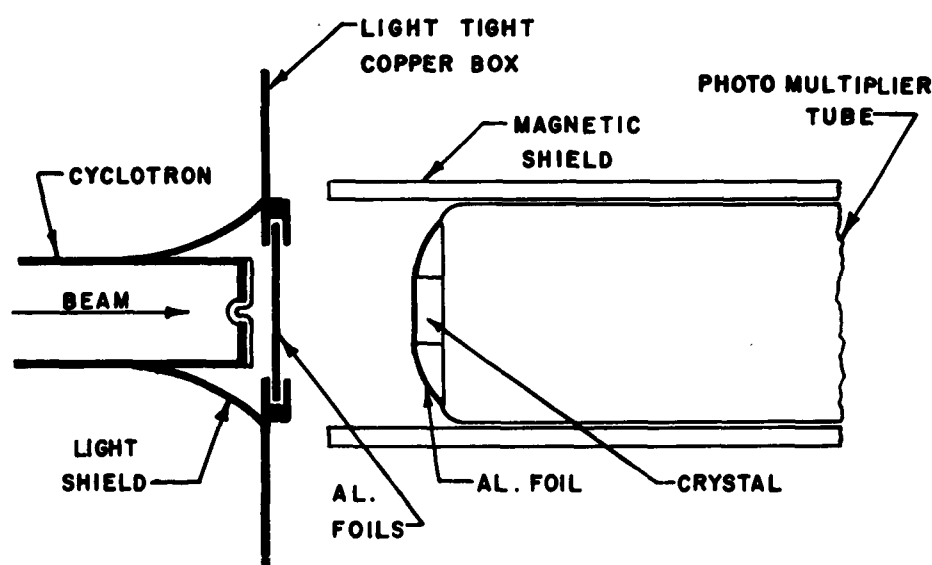


FIGURE 16.-EXPERIMENTAL ARRANGEMENT FOR THE MEASUREMENT OF THE FLUORESCENCE OF CRYSTALS TO HEAVY PARTICLES. NOTE THAT THE DISTANCE FROM THE NYLON FOIL TO ABSORBER AND FROM ABSORBER TO CRYSTAL ARE NOT TO SCALE. TAYLOR, DENTSCHKE, REMLEY, EBY AND KRUGER(46)

The crystals are in optical contact with a photomultiplier tube (type RCA 5819) and wrapped in Al-foil with a thickness of 0.16 mg/cm^2 . Their dimensions are $2.5 \times 2.5 \times 0.3 \text{ cm}$ for anthracene, $1 \times 1 \times 0.5 \text{ cm}$ for stilbene and $1.5 \times 1.5 \times 0.3 \text{ cm}$ for freshly cleaved NaI(Tl). The time elements selected for the photomultiplier output are 0.3×10^{-6} seconds for anthracene and stilbene and 2.5×10^{-6} seconds for NaI(Tl). The results for the anthracene crystal are plotted in Figure 17.

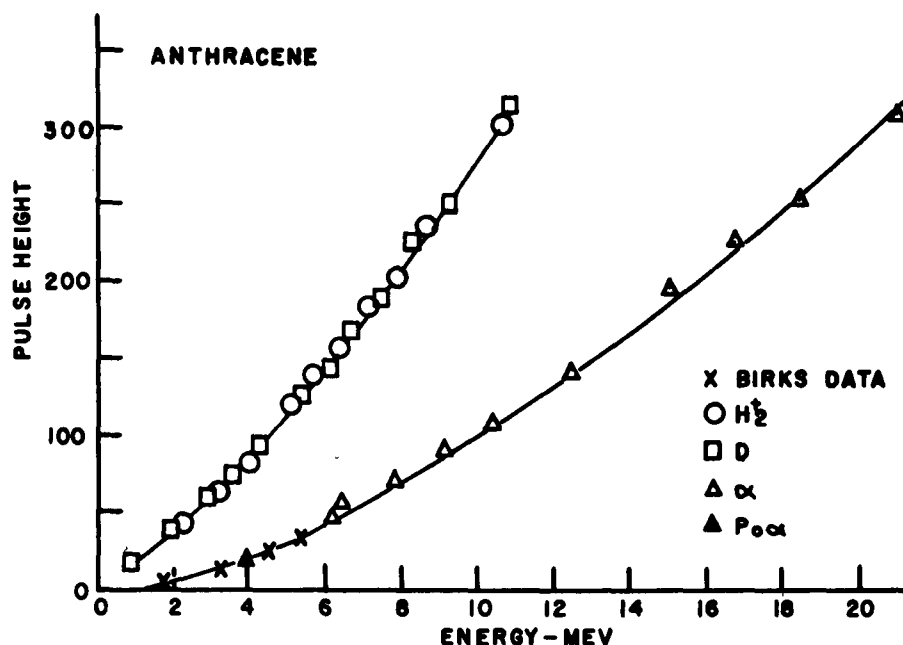


FIGURE 17.-PULSE HEIGHTS RESULTING FROM THE FLUORESCENCE RADIATION OF ANTHRACENE WHEN EXCITED BY HEAVY PARTICLES OF DIFFERENT ENERGIES. TAYLOR ET AL (46)

The organic crystals show no linearity between pulse height and particle energy over the investigated range, while NaI(Tl) gives a linear correlation for protons, deuterons and alpha particles with energies greater than 10 Mev. For alpha particles with energies smaller than 10 Mev Lovberg (47) finds deviations from linearity up to 15%.

The light efficiency-energy function for electrons with energies up to 625 kevis presented in Figure 18. Only NaI(Tl) shows a linear correlation, while the organic scintillators show a deviation from linearity at the energies lower than 80 kev.

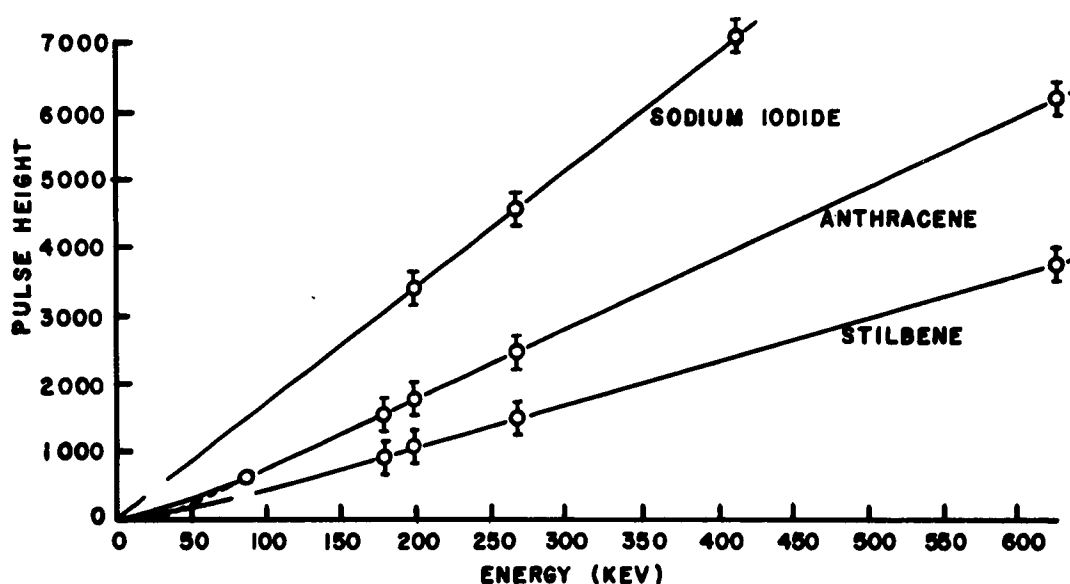


FIGURE 18.—PULSE HEIGHTS FROM FLUORESCENCE RADIATION OF ANTHRACENE, STILBENE, AND SODIUM IODIDE AS A FUNCTION OF ELECTRON ENERGY. TAYLOR ET AL (46)

Hofstadter and McIntyre (48) report proportionality between light output and energy for electrons with energies up to 2.36 Mev.

Measurements of Roser and Bowen (49) make it probable that proportionality between energy and pulse height exists for mesons in the energy range up to 170 Mev.

Efficiency, decay time and spectral emission of a scintillator depend on many factors. Temperature, physical state and foreign substances in the smallest amounts influence and determine the luminescent properties.

The influence of temperature and treatment is manifested in the behavior of naphthalene above and below its melting point. The light output from naphthalene, energized with gamma-rays, slowly drops as the temperature rises from -20° to 60°C , and drops rapidly as the temperature increases from 70° to 90°C . The liquid naphthalene exhibits practically no light emission. Recrystallized naphthalene shows a smaller light emission than powdered material when energized by gamma radiation.

Pure substances and solutions show a dependence of light output and decay time upon temperature as presented in Figures 19 and 20.

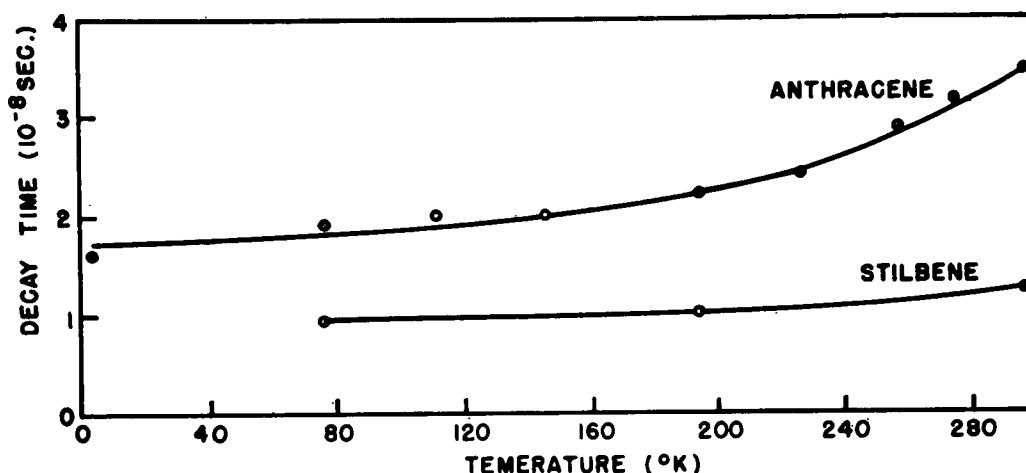


FIGURE 19 - VARIATION OF THE DECAY TIME OF FLUORESCENCE WITH TEMPERATURE. ELLIOT, LIEBSON AND RAVILIOUS (159)

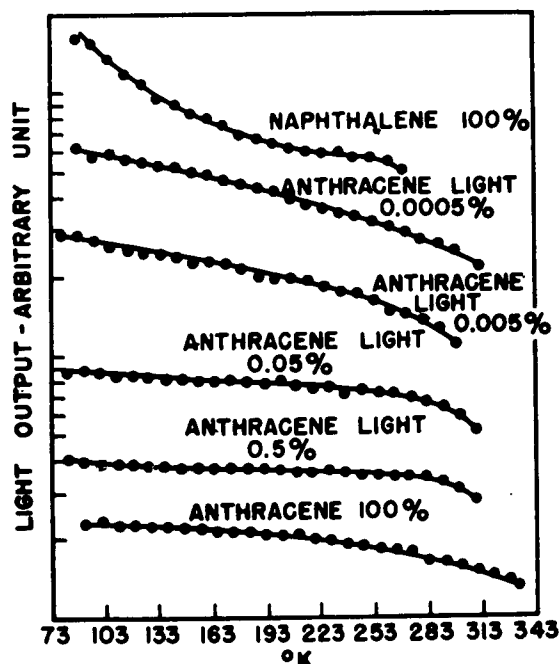


FIGURE 20.- TEMPERATURE CHARACTERISTICS OF NAPHTHALENE-ANTHRACENE MIXTURES. LIEBSON AND FARRAR (162)

Added substances considerably influence the scintillation properties. Small amounts of naphthalene added to anthracene will shift the luminescence spectrum of naphthalene toward the spectrum of anthracene. One anthracene molecule in a thousand naphthalene molecules is sufficient to push the light intensity far into the anthracene region. In solutions, the emission spectrum of the pure substance is broadened and generally shifted to shorter wavelengths, (Table 4).

TABLE 4

Scintillator	Emission Maximum in A°
naphthalene	3300 - 3800
naphthalene in xylene (20 gms/liter)	3200 - 3800
anthracene	4200 - 4700
anthracene in xylene (1.5 gms/liter)	3900 - 4400

Figure 21 demonstrates, according to Kallmann and Furst (50), how terphenyl added to xylene changes the light efficiency conditions in comparison to the addition of anthracene to xylene.

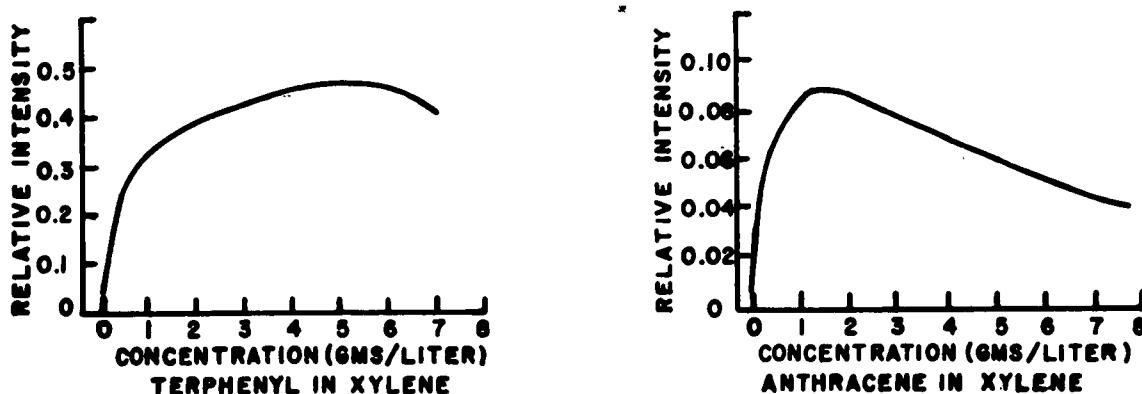


FIGURE 21.- FLOURESCENCE INTENSITY OF DIFFERENT MIXTURES. KALLMANN AND FURST (50)

Foreign substances not only increase the fluorescence intensity (Table 5), but they also are able to quench the light output (Table 6).

TABLE 5
LUMINESCENCE-INCREASING SUBSTANCES IN PURE SOLVENTS.(GAMMA RADIATION. KALLMANN AND FURST (50)).

SOLVENT	SUBSTANCE	CONCENTRATION GMS/LITER	RELATIVE EFFICIENCY *
M-XYLENE			0.012
II	ANILINE	2.	0.036
II	ANTHRACENE	1.4	0.090
II	CARBAZOLE	1.4	0.12
II	TERPHENYL	5.5	0.44
BENZENE			0.011
II	ANTHRACENE	1	0.066
II	CARBAZOLE	1.5	0.074
II	TERPHENYL	4.5	0.28
TOLUENE			0.011
II	ANTHRACENE	1.5	0.074
II	TERPHENYL	5	0.39
HEXANE			0.008
II	ANTHRACENE	1	0.016
II	TERPHENYL	0.8	0.049

TABLE 6
LUMINESCENCE-QUENCHING SUBSTANCES IN PURE SOLVENTS(GAMMA RADIATION. KALLMANN AND FURST(50)).

SOLVENT	SUBSTANCE	CONCENTRATION GMS/LITER	RELATIVE EFFICIENCY *
ANILINE			0.007
II	ANTHRACENE	1.5	0.002
II	CARBAZOLE	0.2	0.005
II	TERPHENYL	0.5	0.005
ETHER			0.006
II	ANTHRACENE		0.004
XYLENE			0.012
II	DIBENZYL	1	0.011
II	DIPHENYL ACETYLENE	1	0.007
II	MELANILINE	1	0.006
WATER DOUBLE DISTILLED			0.009
II	URANYL ACETATE		0.004

* ANTHRACENE CRYSTAL OF THE SAME MASS=1

An amount of 0.1 gm/liter anthracene in a solution of 1 gm/liter terphenyl in xylene decreases the emitted fluorescence light intensity by absorption of the light in the mixture and by internal quenching of the excitation energy. The following data, Table 7, are obtained when gamma radiation is used as the energizing agent (50).

TABLE 7

SCINTILLATOR	CONCENTRATION GMS/LITER	RELATIVE EFFICIENCY*
XYLENE PLUS TERPHENYL	2	0.39
XYLENE PLUS TERPHENYL PLUS ANTHRACENE	0.01	0.21
XYLENE PLUS TERPHENYL PLUS ANTHRACENE	0.05	0.15
XYLENE		0.012
TERPHENYL		0.79
ANTHRACENE		1.0

*ANTHRACENE CRYSTAL OF THE SAME MASS = 1

According to Figure 14, alpha-rays produce stronger scintillations than beta-rays. This, however, does not hold in all cases, as has been shown in Table 2. Kallmann and Furst (50), bombarding solutions with alpha-rays or gamma-rays, found for alpha particles only 20% of the efficiency they obtained with gamma-rays of the same energy. Similar results were reported by Reynolds, Harrison and Hill (51), under the conditions where the amounts of absorbed energy were the same.

These findings, as well as the fact that protons also show a lower efficiency than electrons of the same energy, are explained by a saturation effect related to the higher specific ionization of the heavier particles (Jordan and Bell (52), Frey and co-workers (53), Taylor and co-workers (46)).

It is of interest to follow the interpretation of these findings by Birks (54), Taylor and co-workers (46) and others, who are investigating the relationship between fluorescence light output and ionization density. The importance of the specific ionization or the ion density (number of ions produced per unit path length) for the biologic effects of high energy particles and quanta is well known and has been discussed frequently (Zirkle (55), Tobias (56), Krebs (57)).

A similar importance of the specific ionization for the transformation of particle energy into light energy may be assumed. Special considerations and experiments, however, are necessary for its confirmation.

Taylor and co-workers assume that the differential fluorescence efficiency dL/dE for producing fluorescent radiation is a function of the specific energy loss dE/dx of the particle in the form:

$$dL/dE = e = f(dE/dx)$$

or,

$$dL/dx = e(dE/dx) = (dE/dx)f(dE/dx)$$

The specific energy loss dE/dx is, according to Livingstone and Bethe:

$$-(dE/dx) = (2\pi Z^2 z e^4 N M / m E) \ln(4mE/MI)$$

where Ze , M , and E are charge, mass, and energy of the incident particle. Nz , m , and I are the number of electrons per cm^3 of the stopping substance, the mass of the electron, and the average excitation potential of the atom of the absorber.

N , z , m , and e are constants for a given crystal and the logarithmic term varies between 3.3 and 5.1 for the particles and energies involved. Therefore, in first approximation it can be written, using the constant C : $dE/dx = (MZ^2/E) \cdot C$. This gives: $dL/dE = g(E/MZ^2)$ whence the total light output for an incident particle of energy E is: $L = \int_0^E g(E^1/MZ^2) dE^1 = MZ^2 \int_0^{E/MZ^2} g(E^1/MZ^2) d(E^1/MZ^2)$, or: $L/MZ^2 = h(E/MZ^2)$ where h is a function of E/MZ^2 .

Consequently, for a given crystal the curves of different incident particles should reduce to a common curve, when L/MZ^2 is plotted vs E/MZ^2 . Figure 22 gives the results obtained by Taylor and co-workers (46).

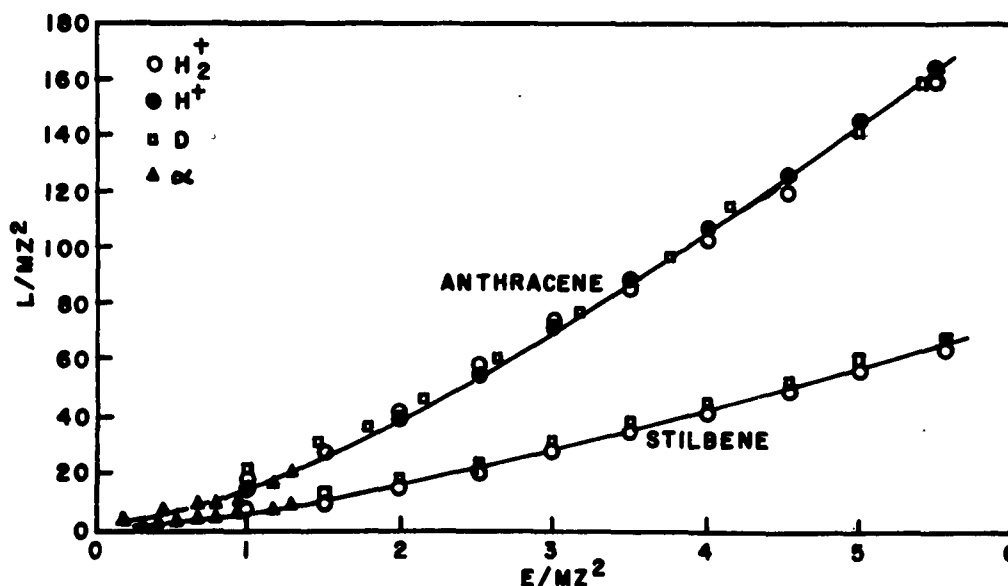


FIGURE 22.- FLUORESCENCE RADIATION OF ANTHRACENE AND STILBENE WHEN EXCITED BY HEAVY PARTICLES OF DIFFERENT ENERGIES WITH ORDINATES AND ABSCISSAS REDUCED BY THE FACTOR $1/MZ^2$; M AND Z ARE MASS AND CHARGE OF THE INCIDENT PARTICLE RESPECTIVELY. TAYLOR ET AL. (46)

For more detailed confirmation, more measurements are necessary, especially with high energy alpha particles. Findings so far confirm the assumption that the fluorescence efficiency depends upon the specific energy loss which in turn is functionally correlated to the specific ionization.

Birks has developed an exciton theory (54) concerning the variation of the specific fluorescence dS/dr with the specific energy loss dE/dr of the exciting particle with an energy E and range r . It leads to the expression: $dS/dr = \frac{A \cdot dE/dr}{1 + kB \cdot dE/dr}$. In this formula $A \cdot dE/dr$ is

the number of excitons produced per unit path length in the organic crystal, $B \cdot dE/dr$ is the local concentration of the molecules damaged by the incident particle, and k is the relative exciton capture probability of a damaged molecule. The values of A and kB are taken from experiments with alpha particles and electrons, being in this case $A = 82.5$ and $kB = 7.15$. This theory holds satisfactorily for the fluorescent response of organic crystals to electrons with energies greater than 20 kev, to protons and deuterons with energies greater than 1 Mev and to alpha particles with energies about 1.6 Mev. For short range particles additional factors must be introduced in the formula as has been done by Birks 1952.

Solid non-crystalline scintillation phosphors have been developed by Schorr and Torney (58), Schorr and Farner (59), Koski (60), and Carlson and Kosky (61). They are solid solutions of terphenyl, anthracene, naphthacene, stilbene and other scintillating substances in organic plastic material. Anthracene and stilbene, dissolved in lucite, paraplex or polystyrene present scintillators comparable to good crystal scintillators. Two percent anthracene or stilbene in polystyrene give pulses for Co^{60} gamma radiation nearly equal to those of single crystals of anthracene or stilbene.

The idea of conducting the scintillation light by means of total reflecting rods over greater distances to the photomultiplier was suggested by Timmerhaus and co-workers (62). The scintillation phosphor may be mounted by these means directly in the beam of a cyclotron (63), in a high pressure apparatus (64), or any place difficult to reach, while the photosensitive detector may be placed at a convenient spot protected against stray magnetic fields, heat, humidity and other disturbing influences. This light piping can be done easily

by lucite rods or quartz rods of proper dimensions. A few values, for illustration, are presented in Table 8. For more details see references.

TABLE 8
LIGHT PIPING WITH A LUCITE ROD 2 CM DIAMETER

SCINTILLATOR - PHOTOMULTIPLIER DIRECT CONTACT	100 % EFFICIENCY
SCINTILLATOR - LUCITE ROD 5 CM LONG - PHOTOMULTIPLIER	85 % EFFICIENCY
SCINTILLATOR - LUCITE ROD 30 CM LONG - PHOTOMULTIPLIER	60 % EFFICIENCY
SCINTILLATOR - LUCITE ROD 120 CM LONG - PHOTOMULTIPLIER	20 % EFFICIENCY

To get ideal geometrical counting conditions Raben and Bloombergen (65) dissolved the radioactive material to be studied in a scintillation liquid. Measurements with C^{14} compounds in a 0.5% terphenyl-xylene solution, faced by two RCA 5819 tubes, allowed the recording of millimicrocuries of C^{14} .

IV. APPLICATIONS

Scintillation counters are used in radioactive investigations as well as for the detection of high energyparticles and quanta. The areas of their application are steadily growing. Measurements of particle energies, gamma-ray spectroscopy, coincidence events, phosphorescence effects, cosmic ray phenomena and radiobiological reactions are a few of the fields in which the counters are used today.

A. Use in Nuclear Physics

The first practical application of a scintillation counter was the measurement of the diffusion velocity of radium emanation and thorium emanation in glass tubes (8). Other applications concern practical technical questions, such as the monitoring of danger areas (66), the recording of atmospheric radioactivity (67) and particle energy determinations mentioned previously. Special devices have been developed for specific problems. Lithium or boron containing phosphors are used for the measurement of slow neutrons. In the case of lithium the scintillations are produced in the alpha reaction, $Li^6(n, \alpha)H^3$ -process. LiF, LiI (Tl), and LiI (SnI_2) proved of special value in these investigations (68). Methyl-, ethyl- or propyl-borates may be added to phenyl-cyclohexane or terphenyl solutions in order to obtain highly efficient neutron counters.

Hydrogen containing phosphors have been used for the detection of fast neutrons. Moulton and Sherwin (69) measured fast neutrons in coincidence arrangement with ZnS-lucite sandwiches. This arrangement gives 30 neutrons per cm^2 per second with a 25 mg Ra-Be source.

Falk, Poss and Yuan (70) used liquid scintillators (organic substances with a high number of H-nuclei per cm^3) for the detection of fast neutrons. Solutions of 0.5% terphenyl in xylene give a very high efficiency for 14 Mev neutrons, however, the light output is not proportional to the neutron energy.

One of the most important and most successful applications of scintillation counters involves the measurement of gamma-ray energies and intensities. This field of beta- and gamma-ray spectroscopy has been highly developed. Scintillation spectrometers with one and two crystals for the measurement of beta and gamma radiations have been described by Bell (71), Jordan and Bell (72), Hofstadter (73), Hofstadter and McIntyre (74), Palmer (75), and others. Jordan and Bell used anthracene crystals; Hofstadter and McIntyre, NaI(Tl) crystals; while Palmer applied anthracene. In these crystals energetic gamma-rays produce photoelectrons, Compton electrons and pairs which appear in the scintillation diagram, depending on their energy and intensity, as pulses of different height and at different locations. Such a spectrogram allows one to draw important conclusions as to the energy of the incident gamma radiation. Figure 23 gives for NaI the absorption coefficient for the photoelectric effect, Compton effect and pair production as a function of the energy, assuming only primary processes occur (74).

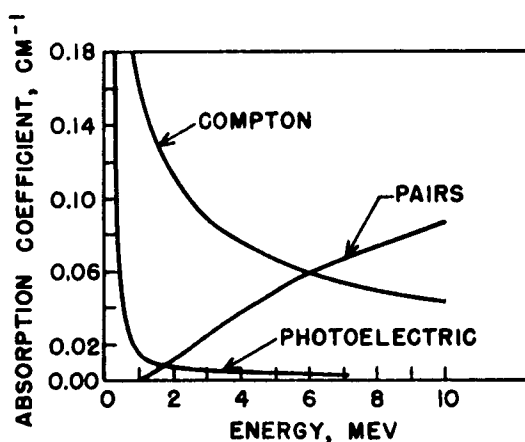


FIGURE 23.- THEORETICAL CROSS-SECTION CURVES FOR PRIMARY PROCESSES IN IODINE OF NaI(Tl). TO INCLUDE THE EFFECT OF SODIUM ADD 21% TO COMPTON CURVE AND 4% TO PAIR CURVE. HOFSTADTER AND McINTYRE (74)

Figure 24 shows a calculated pulse height distribution diagram for NaI under the assumption that 5.1 Mev gamma-rays hit the crystal. Photoelectric, Compton, and pair production lines are clearly distinguished.

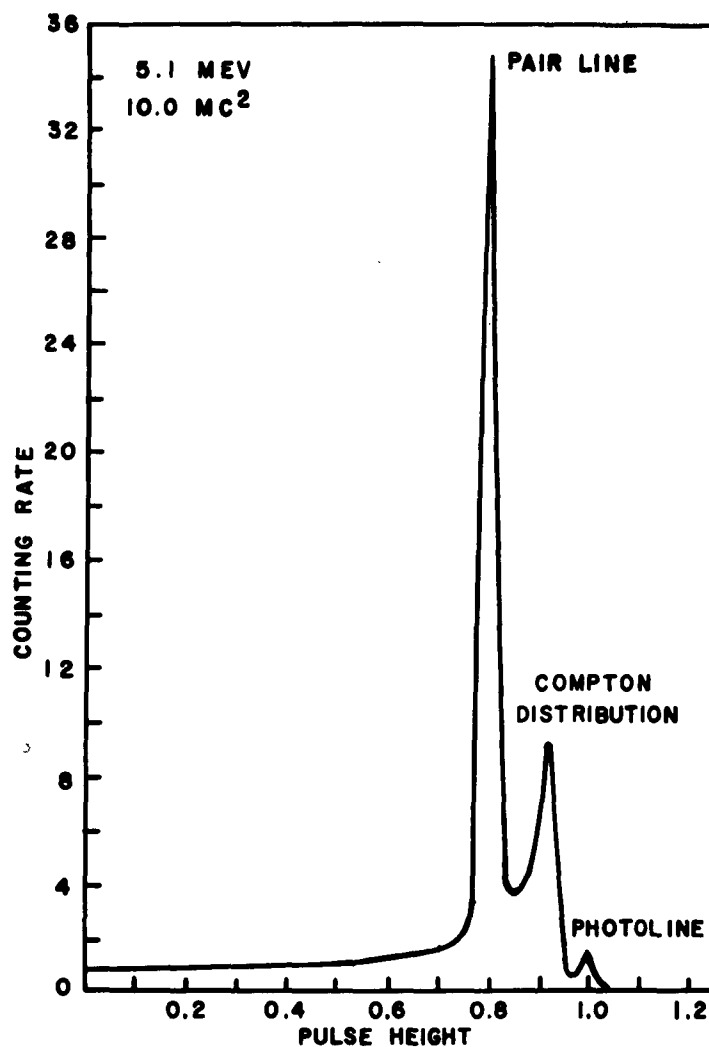


FIGURE 24.-APPROXIMATE THEORETICAL PULSE DISTRIBUTION FOR 5.1 MEV ASSUMING PRIMARY PROCESS ONLY. HOFSTADTER AND McINTYRE (74)

Experiment with Na^{24} gamma radiation with energies of 1.38 Mev and 2.76 Mev confirm the calculations. The pulse height diagram shows the photo-effect line and the Compton-effect line for the 1.38 Mev gamma radiation as well as the other three lines for the 2.76 Mev radiation to be well separated (Figure 25).

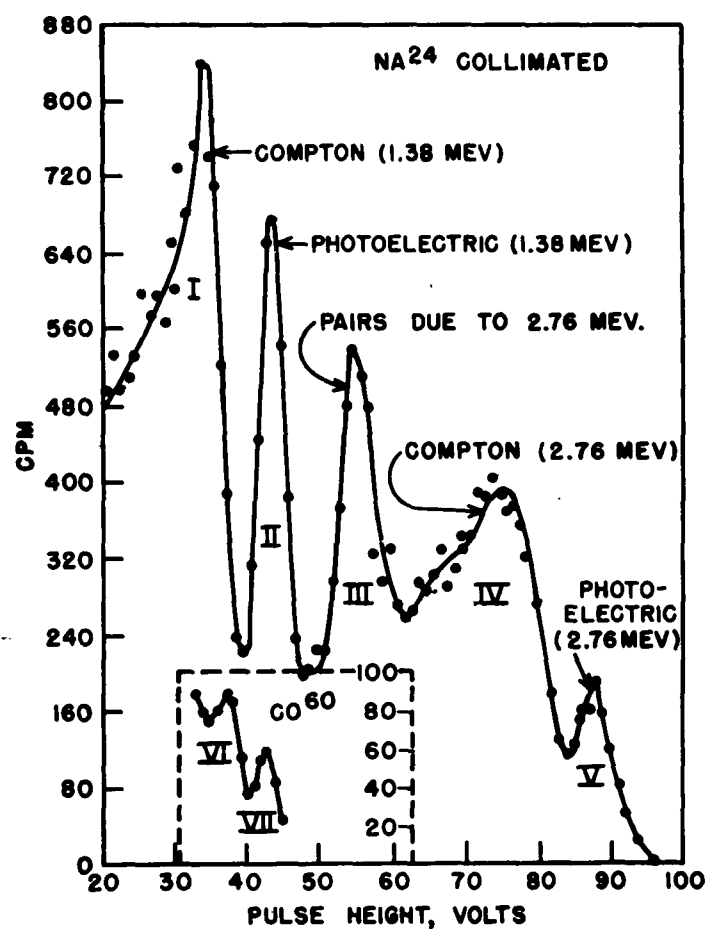


FIGURE 25.- DISCRIMINATOR RUN ON Na^{24} . HOFSTADTER AND McINTYRE (74)

The great sensitivity of NaI(Tl) for gamma radiation was applied by Hammeresh and Hummel (76) for the measurement of neutron capture gamma-ray spectra. Pringle and Isford (77) studied in detail the gamma-rays produced by neutron capture in Mn^{56} and discovered two gamma lines with 5.32 Mev and 7.16 Mev. Maienschein, Blair, and Baker (78) studied the gamma radiation from I^{132} with a special gamma-ray spectroscopy.

The high resolution time of the scintillation counter offers great possibilities for the measurement of very short time intervals and coincidences. Chamberlain, Mozley, Steinberger, and Wiegand (79) measured with stilbene crystals the decay life time of π -mesons. The mesons were produced in a paraffin target by a 340 Mev X-ray beam. They had to pass one stilbene crystal and then came to rest in a second one. The light coming from the crystals was detected by photomultiplier tubes with delayed coincidence circuits. Evaluations of the records give for the meson a mean-life of $(2.65 \pm 0.12) \times 10^{-8}$ seconds, in good agreement with measurements made by other investigators (80).

The half-lives of eight shortlived isomers (Fluegge (81), Segre and Helmholz (82)) were determined by McIntyre (83) using a delayed coincidence method with proportional counters for the detection of the beta radiations and polystyrene crystals for the detection of the gamma-rays. The half-life of Hg^{198} was found to be about 2.3×10^{-8} seconds, while the excited states of Mg^{24} , Hg^{199} , Ca^{42} , Nd^{142} , A^{38} , Te^{122} , and Fe^{56} had half-lives shorter than $(1 \text{ to } 2) \times 10^{-8}$ seconds. The excited states of many nuclei have been determined. McGowan (84) measured 8×10^{-10} seconds for $^{186}\text{Os}_{76}$ and 3×10^{-9} seconds for $^{153}\text{Eu}_{63}$.

Deutsch and co-workers (85) measured the life time of positrons in different gases by using the same principle of delayed coincidences. The mean life time of the positron in nitrogen at atmospheric pressure is 1.5×10^{-7} seconds.

A very impressive application of scintillation counters for measuring time intervals of about 10^{-10} seconds is reported by DeBenedetti and Rickings (86). With scintillation counters in coincidence devices they determined the velocity of gamma-rays over a distance of only three centimeters. Using the positron annihilation radiation of Cu^{64} and Na^{22} (0.5 Mev gamma-rays) Cleland and Sastram (87) measured the velocity of gamma-rays in air over distances up to 30 centimeters. The evaluation of the measurements gives for the velocity of gamma-rays in air the value of $(2.983 \pm 0.015) \times 10^{10}$ cm/sec.

Proton-gamma coincidences in the process $\text{Al}^{27}(\text{d}, \text{p})\text{Si}^{30}$ were studied by Allen, May, and Ball (88) with a NaI(Tl) scintillation spectrometer. They established the decay of Si^{30} .

Many investigations deal with the angular correlation between the particles and quanta emitted in nuclear events (89). Here again the great value of the scintillation counter manifests itself in the fact that without this instrument our present knowledge of these important events would be very limited (40). Stevenson and Deutsch (90) measured the angular correlation in electron gamma-ray emission in radioactive decay with anthracene crystals. Walter, Huber, and Zuenti (91) studied the angular correlation in successive nuclear radiations such as gamma-gamma coincidences, beta-gamma coincidences and electron-electron coincidences.

Special circuits for the observation and the recording of these events have been developed by Meyer, Baldinger, and Huber (92), Shraeder (93), McIntyre (94), Hofstadter and McIntyre (95), and others (96).

The general terminology of luminescence, i. e. of fluorescence and of phosphorescence, is presented in Figure 26 according to the widely discussed scheme of Leverenz (97).

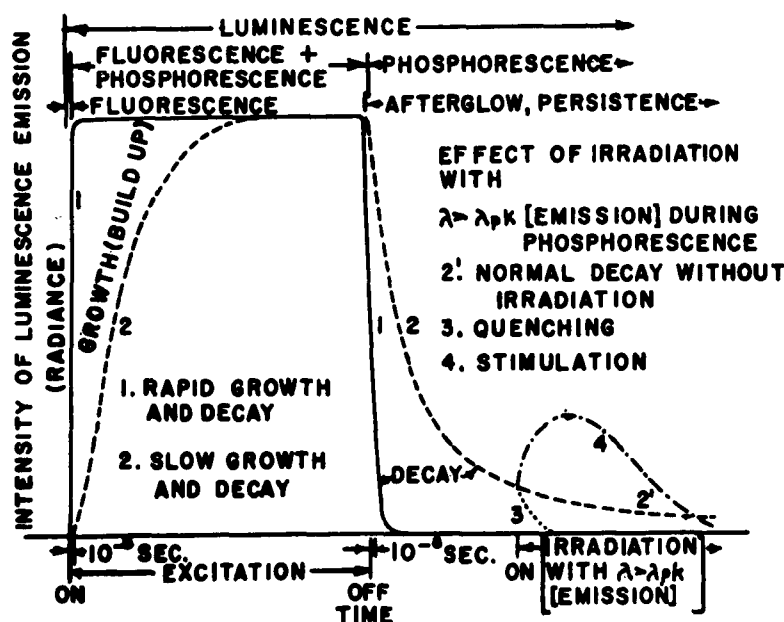


FIGURE 26.- DIAGRAMMATIC REPRESENTATION OF THE DYNAMICS (GROWTH AND DECAY) AND TERMINOLOGY OF LUMINESCENCE EMISSION. LEVERENZ (97)

While fluorescence generally is excited only during irradiation of the phosphor, phosphorescence continues after the irradiation (Figure 26) and can, depending on the kind of phosphor, prevail for long time intervals. This rough distinction (98) has to be appreciated when studying the effect described early by Glasser and Beasley (99). This effect deals with the irradiation of NaCl crystals with X-rays, gamma-rays or high energy particles. Under bombardment with these radiations NaCl exhibits a normal fluorescence. It also shows, however, a strong phosphorescence, the intensity of which can be increased by illumination of the crystal with visible light. Mandeville and Albrecht (100) studied this effect with scintillation counters using silver activated NaCl. They used both types of scintillation counters in combination (101): a photomultiplier scintillation counter for the measurement of the phosphorescence band at about $4,000 \text{ \AA}$ and a phototube scintillation counter for the measurement of the band at about $2,500 \text{ \AA}$. The measurements in the $4,000 \text{ \AA}$ band and in the $2,500 \text{ \AA}$ band make it probable that silver activated NaCl presents a phosphor with two different activators. The band at $2,500 \text{ \AA}$ is due to Ag-Ag pairs in the crystal lattice (Burstein (102), Mandeville and Albrecht (101)), while the band at about $4,000 \text{ \AA}$ is caused by Ag centers in the crystal.

Further systematic studies of this effect are reported by Kallmann and Furst (103) and by Etzel and co-workers (104). They are attempting to explain the mechanisms involved and to find possible practical applications. Since in these cases, as shown by Kallmann and Furst, color centers are formed, the investigations by Friedman and Glover (105) may be taken as a first approach to a practical application of the phenomenon. Friedman and Glover used sensitized KBr crystals for the detection of ionizing radiation and developed a dosimeter on this basis. With 50 r of a 1-Mev gamma radiation they can produce a color change in a crystal of one centimeter thickness, which can be detected with the unaided eye.

B. Use in Cosmic Ray Research

Scintillation counters have been used in cosmic ray research including such important problems as heavy nuclei and explosion stars at the top of the atmosphere. Reynolds (106) reported in 1949 the usefulness of NaI(Tl) crystals for the detection of cosmic radiation and together with Harrison (107) measured the decay of cosmic ray mesons in matter. Williams, Spetner, Kraushaar and Courant (108) built a Wilson cloud chamber triggered by internal scintillation crystals and Salvini and Reynolds (109) studied nuclear explosions with a cloud chamber containing a NaI crystal.

Montgomery and Montgomery (110) recorded cosmic radiations using commercial naphthalene flakes, highly purified anthracene flakes and commercial naphthalene recrystallized from a CS_2 solution.

Harding (111) studied double star effects of the cosmic radiation with KI(Tl) crystals (size $3.1 \times 3.1 \times 1.2$ cm).

Since, in cosmic ray research, areas of several square feet are often necessary, Harrison (112) developed large area liquid scintillation counters. The liquids are in special tanks (see Figure 27) with an effective volume of $10 \times 10 \times 30 \text{ cm}^3$ and are viewed by photomultipliers in a special arrangement.

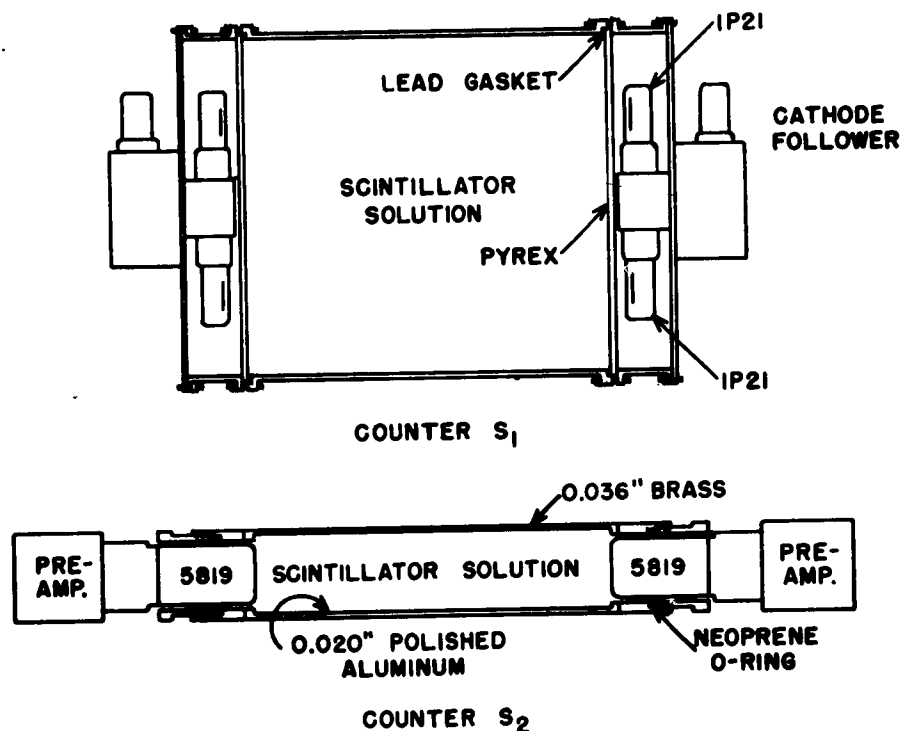


FIGURE 27.- LARGE AREA SCINTILLATION TANK. HARRISON (112)

Ney and Thon (113) used a special scintillation counter arrangement to study the heavy nuclei at the top of the atmosphere. This primary cosmic radiation with its impressive secondary effects in the form of explosion stars has caused many speculations as to its physical nature, its origin and its possible biological effects (114, 115). The experiments of Ney and Thon give, in addition to the photo emulsion studies (116), a welcome contribution to this field. Figure 28 shows the experimental setup they used. Their experiments, done with pyrene-xylene scintillators (8gms/liter), gave results that are in good agreement with the theoretical expectations.

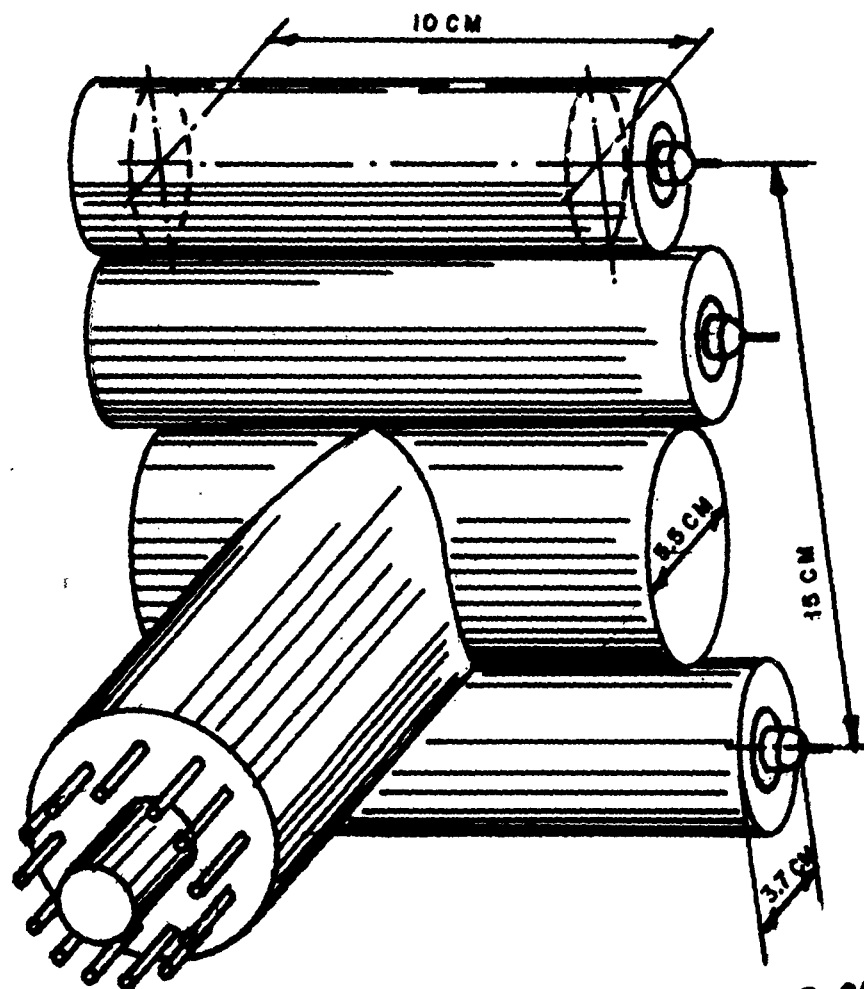


FIGURE 28.—SCINTILLATION COUNTER GEOMETRY FOR COSMIC RAY RESEARCH. NEY AND THON (113)

The distribution of the different components of heavy radiation was studied in a special balloon flight (113). The balloon launched at 55° N. L. was at an altitude of 105,000 feet for five hours and recorded the following pulses, (Table 9).

TABLE 9

ELEMENT	SCINTILLATION COUNTER		PHOTO EMULSION	
	NUMBER OF PULSES	PERCENTAGE	FLUX PER CM ² SEC STERAD	PERCENTAGE
H	8740	82	1900	84
HE	257	11.8	280	12
LI, BE, B, STARS	387	3.6	84	3.7
C, N, O, STARS	76	0.7	16	0.7
Z > 9, STARS	20	0.2	4.3	0.2

Recent investigations by Lal, Dal, Kaplon and Peters (117) on the composition and the variation of the primary cosmic radiation with time confirm these findings, however, they place greater emphasis on the heavier groups C, N, O, and nuclei with $Z > 10$.

C. Use in Radiobiology and Medicine

Radiobiology and medicine also have taken advantage of the outstanding properties of the scintillation counter (118). So far, dose measurements, determination of isodose curves, tracer studies with different isotopes and clinical radioactive investigations are the main fields. New possibilities are offered by the high gamma-ray sensitivity of the counter and its short resolving times.

Reiffel and Burgwald (119) measured with anthracene crystals beta- and gamma-ray intensities in the energy range from 10^{-6} r per hour up to 100 r per hour. Great stability, high sensitivity, small dimensions and handiness are desirable, thus the advantage of using the counter. Webb, Paul and Dart (120) used wurtzite for the measurement of X-rays with wavelengths from 5.6 Å to 15.5 Å (800 to 2200 V) and demonstrated with this scintillator the Al-K line at 8.32 Å. West, Meyerhof and Hofstadter (121) as well as Neissel and Moore (122) measured X-rays with NaI(Tl) crystals.

The equivalence between air and tissue dose for gamma-rays was studied by Ter-Pogossian, Ittner and Aly (123), using scintillation

counters and a water phantom. In contrast to the earlier measurements with electron metal ionization chambers by Griffith (124), they found the ratio of air dose to tissue dose to be greater than one for volumes between 10 and 60 cm³. More detailed measurements are necessary for the explanation of this discrepancy.

These measurements are closely connected with the possibility of using solid scintillators for dose measurements in place of the presently used gas ionization chambers. This would provide a dose unit which would hold for all ionizing radiations. In this case the scintillator must be a substance whose photo-electron and Compton-electron emission is equivalent to that of air. An important step in this direction was taken by Ittner and Ter-Pogossian as well as by Breitling and Glocker (125) who studied the wavelength dependence of scintillation counters for X-rays over a wide energy range. Anthracene showed in the studies of Breitling and Glocker a strong wavelength dependence as is apparent from Figure 29. The slope of the curve in the short wavelength range agrees with measurements by Smeltzer (126) for voltages between 250 and 1200 kev.

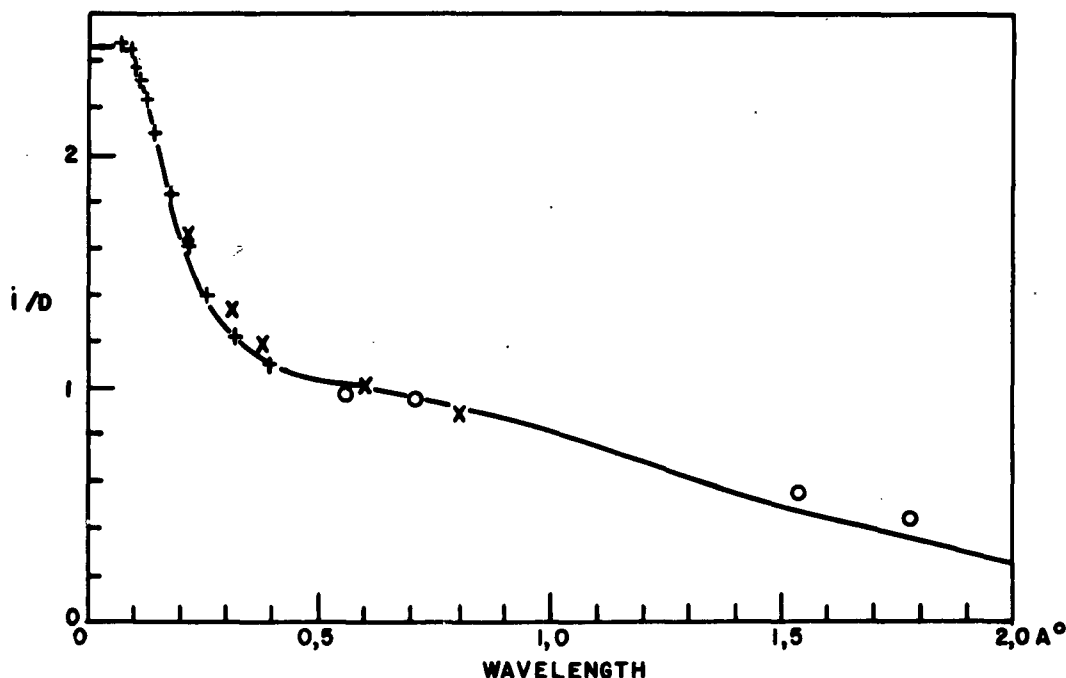


FIGURE 29.-WAVELENGTH DEPENDENCE OF SCINTILLATION COUNTER (ANTHRACENE) FOR X-RAYS. BREITLING AND GLOCKER (125)

Liquid scintillators also show a strong wavelength dependence. Mixtures of solutions in the proper concentration have proved to be independent of the wavelength within certain wavelength ranges. Thus, 0.5% terphenyl in 0.2% chlorbenzene plus 0.8% benzene solution are practically wavelength independent over the wavelength region of 0.08 to 0.6 μ , (Figure 30).

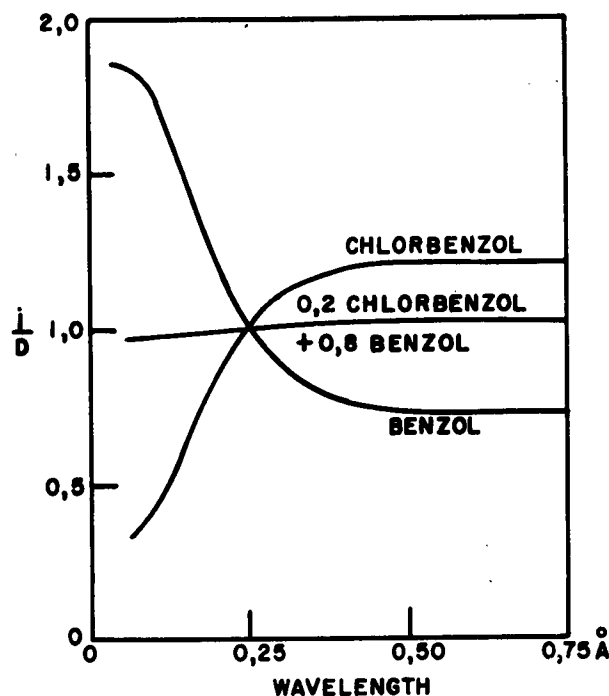


FIGURE 30.- WAVELENGTH DEPENDENCE OF TERPHENYL IN DIFFERENT SOLUTIONS. BREITLING AND GLOCKER (125)

Since solid scintillators fit the requirements better than liquids, mixtures of two crystalline substances were investigated. Mixtures of phenanthrene and chlorphenanthrene satisfactorily matched the requirements for wavelength independence.

Isodose curves of clinically used radioactive preparations were taken by Hine, Berman, and Elkind (127). The equipment is completely automatic and satisfies practical needs.

Scintillation arrangements for the location of incorporated isotopes in clinical studies have been developed recently (128). Kohl (129) built a scintillation needle for the localization of brain tumors. The scintillation crystal with a volume of 6 mm^3 is fixed at the tip of a needle which is 8 cm. long and 3 mm. in diameter. The bore of the needle is highly polished and filled with a lucite rod for light piping. Stilbene proved to be better than anthracene, CdWO_4 or CaWO_4 for these studies.

Kohl, Moore and Chou (130) developed scintillation counters for general clinical work with radioactive isotopes, while Corbett and Honour (131) constructed a directional counter for thyroid studies.

The fate of incorporated radioactive substances in the organism, their influence on different metabolic and physiological processes and their distribution in the different organs and tissues are the main problems of radium therapy and related fields. Beta- and gamma-ray emitting substances can be traced in the living intact organism relatively easily. The measurement of soft radiation emitting substances in the living organism is, however, an unsolved task. Here the scintillation counter offers great possibilities. Cassen and Curtis (132) made in vivo measurements on animals with a cadmium sulfide-phosphor using long lucite rods and a photomultiplier tube type 1P21. The photon tube scintillation counters with their high area sensitivity also (26) should be well fitted for this purpose. Dwarf photon counters will be of special value for small volume investigations.

Herforth and Schaefer (133) applied the scintillation counter in special work in surgery. To check the revascularisation of skin transplants, they injected uranylacetate solutions and observed the uptake of this alpha emitting substance by the transplant. The comparison of the number of emitted alpha particles in normal skin and in the transplant was considered a measure for the revascularisation of the transplant.

A new field of research with radioactive tracers takes advantage of the fact that scintillation counters are several hundred times more sensitive to gamma radiation than any other gamma-ray detecting device. Thus, gamma-ray emitting isotopes can be applied in the future on a similar, perhaps even broader, scale than the presently used beta-ray emitting substances.

Photomultipliers and scintillators have been introduced also into the pioneer investigations dealing with the detection of Čerenkov radiation - a radiation produced when particles with very high energies are stopped in matter. Marshall (134) developed a Čerenkov radiation

counter for fast electrons. Mather (135) applying photographic techniques measured the energy and velocity of high energetic protons and Marshall (136) and Birge (137) discussed the general possibilities.

V. FUTURE DEVELOPMENT

New photomultiplier designs, improved photon counters, better phosphors, high speed circuits and oscilloscopes, pulsed operation of multipliers, understanding and explanation of the fundamental processes occurring in fluorescence and phosphorescence under bombardment with high energetic particles and quanta are the main areas for future development.

Photomultipliers with especially large photocathodes and extremely high gains have been developed by Greenblatt, Green, Davison, and Morton (138). The photomultiplier tube RCA H-5037 has a photocathode with a diameter of 3.5 inches and gives, with 10 dynodes and an overall voltage of 1,000 volts, a gain of 10^8 . The electrons produced in the photocathode are focused onto the first stage of the multiplier by a cylindrical electrostatic lens. The tube type RCA 4646 has 16 dynodes, providing a gain of 10^9 with an overall voltage of 2,000 volts. The resolution time of these tubes is 10^{-8} to 10^{-10} seconds and the sensitivity 30 to 50 ua/lumen. The English tube type EMI 6262 has 14 dynodes, a photocathode with a 5 cm diameter and an amplification factor of 10^9 .

Special high speed and coincidence circuits have been developed by Marshall (139), by Bay, Meijer and Papp (140) by Wouters (141) and by Wells (142). Marshall used a 6BN6 gated-beam tube (Figure 31) and obtained resolving times better than 3×10^{-10} seconds.

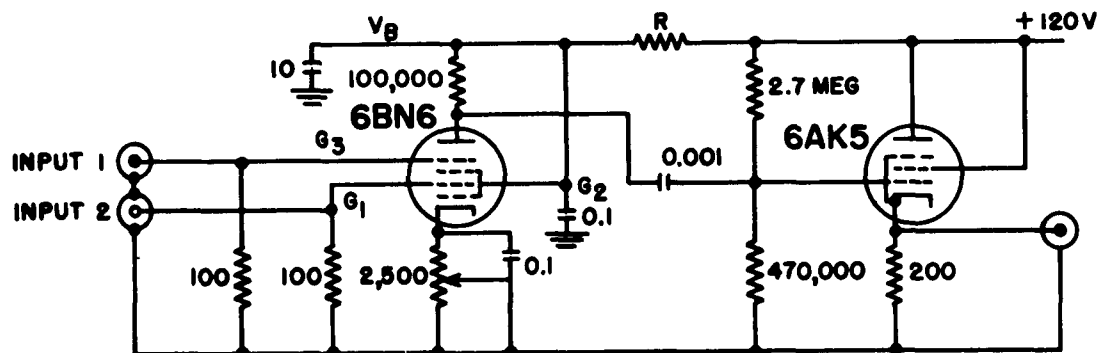


FIGURE 31.- FAST COINCIDENCE CIRCUIT USING 6BN6 GATED-BEAM TUBE.
FISCHER AND MARSHALL (139)

Bay and co-workers applied germanium diodes in a differential, fast coincidence-anticoincidence circuit, getting resolving times of about 2×10^{-10} seconds. Considerations by Post (143) and by Morton (144) show that these values are close to the highest possible resolution time of the best scintillation counters, which may be close to 4×10^{-11} seconds for stilbene. A high speed oscilloscope with 10^{11} sweeps per second has been built by Smith (144).

Attention must be given to the satellite pulses and the afterpulsing (145). Proper construction of the multiplier, enforced gas exhaust and the application of silver-magnesium dynode surfaces may suppress these effects.

Improved photosensitive Geiger counters have been constructed by Mandeville (146) and others (147, 148). With special photocathodes and special circuits it is possible to get resolution times of 10^{-6} seconds and proper counting rates of 200,000 counts per second. Phototube scintillation counters, therefore, may prove of advantage in many cases where a 360° solid angle of detection is desirable. Figure 32 shows a dipping scintillation counter, developed by Parr and Krebs (149), which permits the application of large scintillation volumes and extended gamma-ray sources.

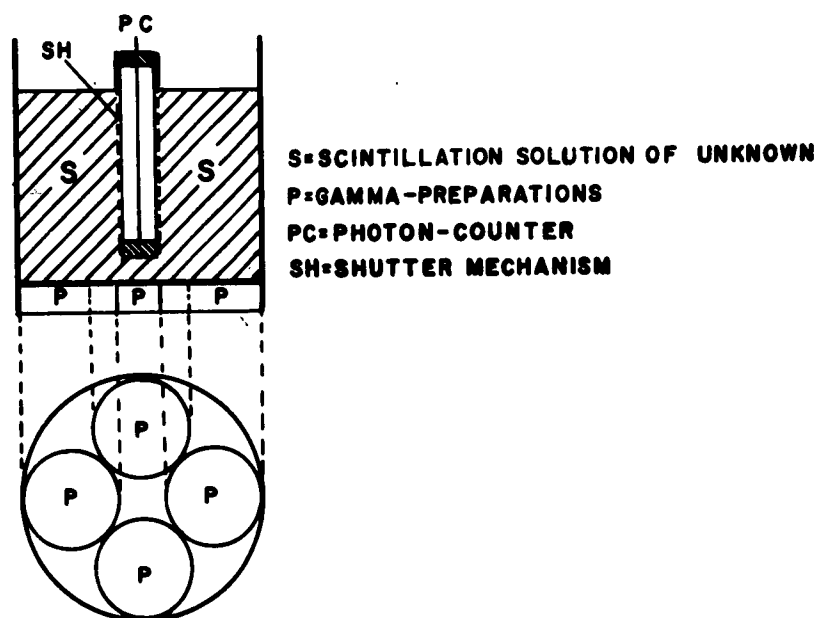


FIGURE 32.- SCINTILLATION DIPPING COUNTER (149) (SCHEMATIC)

The combination of both types of scintillation counters, first tried by Mandeville and Albrecht proved very valuable. A kind of combined scintillation counter suggested by Ramsey (15) is shown in Figure 33.

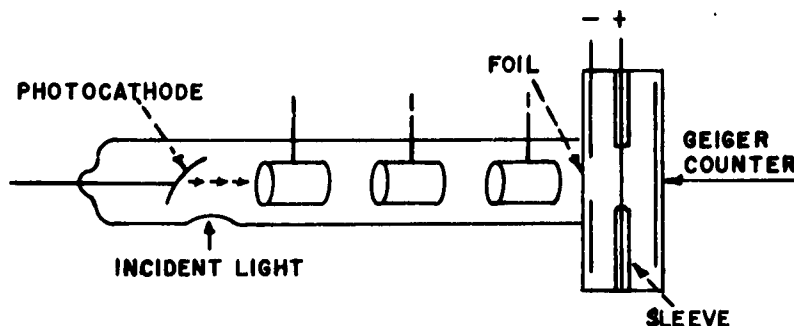


FIGURE 33.- PHOTSENSITIVE COUNTING DEVICE SUGGESTED BY RAMSEY (15)

Much emphasis is being placed on the discovery and production of new phosphors. During the third symposium on scintillation counters (150) the following reports were made: Reynolds, on general properties of scintillation phosphors; Schenk, on different lithium compounds for neutron detection; Bernstein and Schardt, on studies with activated lithium crystal detectors; Swank, on investigations of borate-phenyl-cyclohexane solutions as a means for the detection of slow neutrons; and Harding, on plastic phosphors.

In addition to the interest in new experimental devices, developments and applications, the present tendency of scintillation research aims at an understanding of the fundamental processes involved in scintillation substances when bombarded with high energy quanta and particles. According to present theories scintillations in crystalline phosphors with foreign atoms are produced by an energy transport of the incident energy from the place of incidence to the place where the foreign atoms are located in the crystal lattice. The foreign atoms absorb this energy and usually emit it in the form of light energy (97, 98). This process of energy migration has been discussed extensively by Franck and Livingstone (151), Pringsheim (98), and Leverenz (97). Therefore, science has today a fairly clear picture of certain of the involved mechanisms. However, little is known about the processes involved in fluorescent organic solids and in liquids. The investigations of liquids under high energy particle and quanta bombardment have given new hints in this direction. Hays and Liebson (150) using compounds containing C^{14} and H^3 investigated the fundamental processes of energy transfer in organic fluors. The evidence

obtained so far is consistent with the theory that the initial ionization steps are followed by an energy transfer from solvent cations to solute molecules. In connection with this process, the produced solute cations recombine with low energy electrons to give excited scintillator molecules subsequently returning to their ground states with a simultaneous emission of light.

Breitling and Glocker (125) as well as Kallmann and Furst (50) have suggested further possible mechanisms for liquids. According to Kallmann and Furst the incident energy is taken up by the molecules of the bulk and from there transported to the foreign substance molecules where it is radiated or quenched depending on the kind of molecule.

The problem of energy migration in scintillating substances, also discussed by Reynolds (152) is of interest and importance to radiobiology as well as physics. For years radiobiologists have been discussing the way in which the incident ionizing energy is transported to the place where it produces its biological effect (153). The knowledge gained from work with crystalline phosphors and dye solutions (154) has been evaluated and applied extensively in the field of radiobiology. Butenandt, Dehlinger, Dorneich, Friedrich-Freksa, Krebs, Langendorff, Moeglich, Rajewsky, Rompe, Schaefer, Scheibe, Sommermeyer, Timofeeff-Ressovsky, Zimmer and others made important contributions to this problem during the Radiation Symposium at Radiumspa Oberschlema (Radium Research Institute of the Kaiser Wilhelm Society) in 1941 (173).

The same question arises now (Kallmann and Furst (155)) in connection with new investigations of the scintillation properties of solids and liquids. In this case the application of the scintillation counter with its high resolution has provided new information concerning the stimulation and quenching processes of different materials. It is now well known that in liquids and solutions the incoming energy can be re-radiated by the foreign substance molecules as luminescence light. It is also known that in certain cases the incident energy can be quenched, preferably by the foreign substance molecules and converted into heat or trapped and possibly utilized in a chemical reaction.

According to the newer ideas presented by Kallmann and Furst (155) it is certainly possible that similar processes may occur in other systems, for instance in a biological system. The incident energy, taken up by the added foreign substance molecules, is then utilized in a heat reaction, in a chemical reaction, or in other possible ways.

With these facts in mind it is of interest to discuss the protective effects of cysteine and glutathione against X-irradiation when given prior to exposure. What provides this protection is at present unknown.

Different possibilities have been suggested and discussed. They concern the caging of activated water products by the added substance, the protection of enzymatic activity, the interference with important metabolic cycles such as the Krebs cycle, the nucleic acid cycle and, as recently discussed by Brues and Patt (156), a possible simple "shielding effect" of the incorporated cysteine and glutathione.

For the latter case the presented considerations offer a welcome support. Assuming that cysteine may play the role of the foreign substance in the biological system as does terphenyl in xylene or toluene, it may be possible that the incident X-ray energy is absorbed by the molecules of the bulk material and from there as postulated in one of the suggested mechanisms (155), transported to the cysteine molecules which in turn use up the energy in a manner harmless to the biological system. In such a case a part of the incident X-ray energy would be used up by the added protective substance allowing for a considerable increase in the LD₅₀ dose. It may be mentioned that many intensity-concentration curves for mixed solutions, as given by Kallmann and Furst, show a similarity to the cysteine protection-concentration curve of Patt and co-workers (157).

It is very tempting to study the mentioned possibilities in relation to ideas such as activated water products, inhibition of enzyme activity, interference with metabolic cycles, shielding effects and related processes. Since biological systems are, however, much more complicated than the simple systems that have been investigated, more detailed studies are necessary in order to check whether or not the new insights gained from work with scintillation liquids can be applied to radiobiological problems.

A promising tool for many of these studies is the scintillation counter with its outstanding properties.

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